ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, U. S. ATOMIC ENERGY COMMISSION,
By ISOTOPES INFORMATION CENTER, OAK RIDGE NATIONAL LABORATORY

Fall 1969

● VOLUME 7

● NUMBER 1

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ISOTOPES INFORMATION CENTER

Isotopes and Radiation Technology

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Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 20402, $2.50 per year.

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

Insofar as feasible all references are checked by the editors, using the library facilities of Oak Ridge National Laboratory, Oak Ridge Associated Universities, and the University of Tennessee. Occasionally, however, the cited material is not available; such cases are identified by an asterisk (*).

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

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OAK RIDGE NATIONAL LABORATORY

Issued quarterly by the U. S. Atomic Energy Commission. Use of funds for printing this publication approved by the Director of the Bureau of the Budget on December 16, 1968.

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Section

I

Isotopes and Radiation Technology

Isotope Production and Development

ORNL Isotopes Target Program, 1965-1967*

By E. H. Kobisk†

Abstract: *The facilities and capabilities of the Oak Ridge National Laboratory Isotope Target Center are described. Reduction–distillation and crystal-bar techniques for preparing high-purity materials are discussed, as are various procedures for preparing self-supported and supported films by vacuum evaporation and for making rolled foils.*

The Oak Ridge National Laboratory (ORNL) Isotope Target Center was described previously[1] in this publication. Since that time research, development, and production activities of the Center have expanded greatly to meet the increasing number of service requests. Technology unique to the preparation of thin film targets has broadened and has been applied to the formation of ultrapure-isotope and normal-assay materials for use in nuclear interaction research and other programs more closely associated with solid-state physics and metallurgy. In addition, more emphasis has been placed on fabrication of targets containing the actinide elements. In 1964 the Center produced an average of 100 targets per month; in 1967 this average had increased to 250. Many additional samples (e.g., plates, castings, wires) which, strictly speaking, cannot be classified as targets, were prepared for a variety of research programs other than nuclear physics.

High-Purity Isotope Materials Program

Contaminants often result in significant changes in the physical and chemical properties of elemental isotopes used in target preparation and can reduce the accuracy and sensitivity of results in experiments utilizing the targets. The effect can be particularly detrimental in the preparation of thin, self-supporting films.

Traditional purification techniques are usually inadequate for use with enriched isotopes because of low product yields or because of the possibility of degradation of isotopic purity by contamination with normal material. Therefore methods for purifying milligram to multigram quantities of isotopically enriched material are required. A number of purification techniques are used at the ORNL Isotope Target Center.

Reduction–Distillation of Metals

The reduction–distillation technique is applicable to the preparation and purification of many isotopes in the metallic state.[2] Four critical characteristics of the metal reductant are necessary to achieve successful reduction–distillation:

1. The reductant metal must react with the metal oxide at a temperature which is not too different from that at which the product metal exhibits a vapor pressure of ~1.0 torr.
2. The reductant metal should be close to or above its melting point at the desired reaction temperature.
3. The vapor pressure of the reductant metal must be negligible ($<10^{-7}$ torr) at the reaction temperature.
4. The reductant metal oxide(s) must have a vapor pressure that does not exceed or (preferably) is less than that of the metallic form.

Most metal reductants can be excluded on the basis of the vapor pressures at the reaction temperature,

*This is an edited version of ORNL-4308, Review of Isotopes Target Program, January 1965–December 1967, October 1968.

†Isotopes Development Center, Oak Ridge National Laboratory.

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which itself depends on the metal oxide to be reduced. The reductants most often used are Al, La (and other rare earths), Zr(Hf), Ta, Th, and, upon occasion, U.

The preparation of magnesium metal illustrates the dependence of the reduction–distillation method on the characteristics of the reductant. Although aluminum metal reduced MgO, the yield of product metal from this reaction was invariably low (usually 50 to 60% of the theoretical amount), and aluminum contamination in the product ranged between 200 and 1000 ppm, depending upon temperature control. It was found with lanthanum metal that product yields increased to 90 to 95% since a higher reaction temperature (~1150°C) could be used. Furthermore, lanthanum contamination of the magnesium never exceeded 50 ppm.

Most reduction–distillation reactions proceed smoothly, and, when the reductant metal is above its melting point, the rate of product vaporization (reaction) can be easily controlled by varying the temperature. However, several reductants (e.g., zirconium and tantalum powders) cause reduction well below their melting points. Tungsten metal also has this capability, but the reaction product, WO_x, is highly volatile and causes gross contamination. With each of these three "solid" reductants, there appears to be a very distinct temperature at which rapid reduction–volatilization occurs and below which no reaction is observed. The rate of volatilization is difficult to control since reduction does not occur until a temperature has been reached that is several hundred degrees above that required to produce a vapor pressure of ~0.1 torr, the pressure usually employed in vacuum distillation–condensation procedures.

The reduction–distillation technique has been used to prepare elemental high-purity isotopes of potassium, all alkaline-earth (Group II-A) elements except radium, and all lanthanide elements except lanthanum and cerium. This process is used only for those metals which cannot be obtained in quantity as efficiently or in sufficient purity by some alternative method. Table 1 summarizes the conditions and efficiencies of the reduction–distillation technique for those elements whose isotopes are now being made available as metals by this technique.

Table 1 Conditions for Metal Isotope Preparation by the Reduction–Distillation Technique

Metal oxide	Reductant	Reaction temp., °C	Crucible material	Heating technique*	Collection efficiency, %
Mg	La	1050–1100	Ta	R	87
Ca	La	1150–1200	Ta	R	85
Sr	La	1025–1075	Ta	R	85
Ba	La	1150–1250	Ta	R	58
Si	Ta		C	EB	–
Nd	Th	1950–2000	Ta	R	75
Pm	Zr	1500–1700	W	EB	–
Sm	La	1300–1350	Ta	R	78
Eu	La	1150–1200	Ta	R	92
Gd	Th	1900–1950	Ta	R	63
Dy	Th	1750–1800	Ta	R	72
Er	Th	1850–1900	Ta	R	85
Yb	La	1125–1200	Ta	R	75
Np	Zr	~1400	W	EB	–
Am	Zr	1000–1100	Ta	EB	50

*R, resistance (ohmic) heating; EB, electron-bombardment heating.

Elemental calcium isotopes were initially prepared for accelerator and solid-state-physics research by reduction of CaO with lanthanum metal and subsequent (or simultaneous) distillation of the product metal from the cermet compact. When large quantities (1 to 100 g) of metal were required, a modification of the process was found applicable—"fractionation" of metal vapors. By using a distillation column integrated with and above the reaction zone, as illustrated in Fig. 1, higher reaction temperatures could be employed with associated increased yield of product metal. The

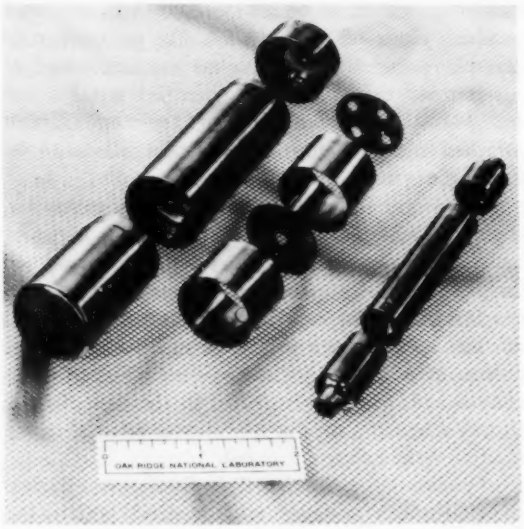

Fig. 1 Metal reduction–distillation column. (a) Top section is effusion port, center section is distillation column containing perforated plates, and lower section is reaction vessel. (b) Disassembly of distillation-column components. (c) Similar still used for <0.5-g quantities of material.

perforated plates in the column were maintained at a temperature lower than that of the reaction zone so that impurity metals with vapor pressures lower than that of calcium were condensed, and increased purity of the product was achieved. The requisite temperature gradient up the column was created by varying the number and spacing of the helical induction coils surrounding the reaction–distillation vessel. Figure 2 illustrates this technique as applied to the preparation of 65 g of ^{40}Ca metal. A total impurity content of <300 ppm by weight was achieved with a product yield of 95%. Eventually this product metal was pressed into 1-in.-diameter pellets for neutron-scattering experiments.

The reduction–distillation method can also be used to separate a specific element from a mixture of elements. A combination of Eu, Gd, and Sm in the ratio 100 : 20 : 5, respectively, was fractionated in a tantalum column. After a single reduction–fractionation, the purity of the europium was >99.5%. The gadolinium and samarium components were retained in the reaction mixture. Similarly the purity of some actinides can be enhanced by application of this technique, which is particularly useful where fission products accompany the desired element.

Application of the fractionation technique in the preparation of ultrahigh-purity ^{151}Eu and ^{153}Eu metals permitted the fabrication of elliptical targets weighing 10 g each and having major and minor axis dimensions of 3.25 and 2.25 in., respectively. These targets were cold rolled in a purified argon atmosphere to inhibit oxidation.

Other high-purity lanthanide metals were prepared by the reduction–fractionation technique. Cold rolling of various isotope or normal metals derived in this manner resulted in foils with a minimum areal density of ~800 $\mu g/cm^2$ (0.0008 to 0.0012 in.) for an area >6 cm^2 and ~500 $\mu g/cm^2$ for ~2-cm^2 area. The total oxygen and nitrogen contents of these metals were usually <500 ppm.

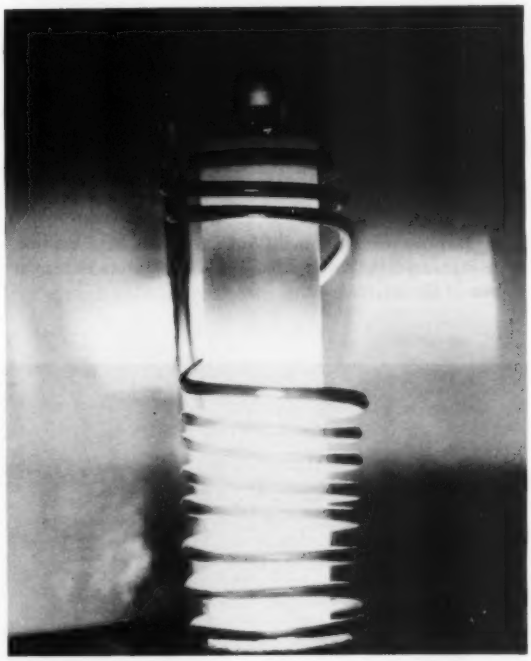

Fig. 2 Reduction–distillation of isotopically separated ^{40}Ca in an induction-heated tantalum column. Differential heating of the column allows purification by fractional distillation. Ionized calcium metal vapors are issuing from the top of the column.

van Arkel–de Boer Purification of Isotopes

The van Arkel–de Boer crystal-bar purification process[3] has been used commercially to obtain high-purity Group IV-B metals. Poor yields (~20 to 50%) usually result in the manufacturing-scale process; however, a modified process has been developed for small

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Fig. 3 Hot-filament portion of van Arkel–de Boer reactor. Crude titanium-metal sponge and iodine crystals can be seen at the base of the reactor.

quantities of these metals without loss and with product yields of ~75 to 95%. For purification of isotopic material, a Pyrex glass reaction vessel of ~1-liter capacity is used. The purified metal is collected on an 0.008-in.-diameter tungsten filament attached to the two tungsten electrodes sealed into the top of the vessel.

In operation the impure metal, usually as powder, is poured into the vessel to a level which is out of contact with the tungsten electrodes. Powdered samples have high surface area and considerable sorbed gas that can cause contamination and loss of metal ductility by being sorbed in the crystal bar during the process. Hence, long outgassing periods are required to desorb and remove the bulk of these impurities. Usually a vacuum of 2×10^{-9} torr is attained in the reaction vessel before it is sealed.

Addition of purified iodine from a sealed glass ampule (broken inside the reaction vessel) initiates the purification process. The entire sealed vessel is placed in a furnace and maintained at the appropriate iodine–metal reaction temperature. The tungsten filament is heated to the required temperature, and thermal deposition proceeds until no further growth is evident. Figures 3 and 4 illustrate the growth process and the resultant bars of material obtained by this technique.

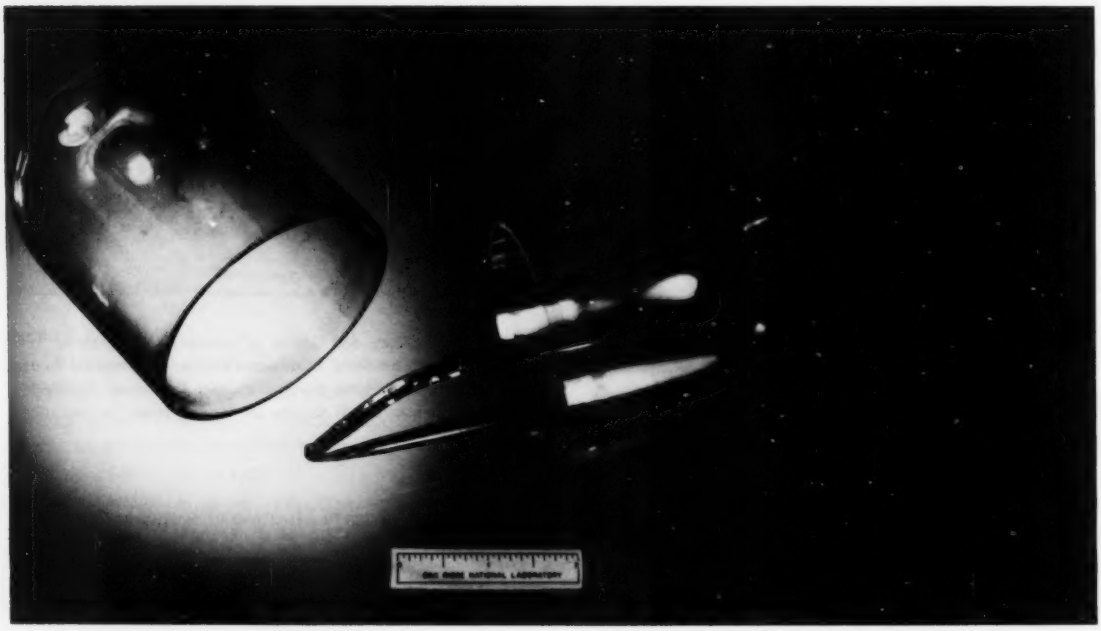

Fig. 4 Crystal-bar zirconium metal as formed on the tungsten filament.

When a crystal bar is employed as starting material for mechanical rolling of thin foils, the metal is cracked from the tungsten filament, and the remaining pieces of tungsten are removed under a microscope. Unfortunately, the layer of metal surrounding the filament is usually highly contaminated with tungsten to an unknown depth, and the tungsten is generally not removable from the bulk of the material. When titanium is being prepared, the tungsten filament is completely dissolved by the resulting crystal bar.

Two techniques were developed to eliminate tungsten contamination: (1) use of increased filament temperature to form an annular void around the filament and thus prevent tungsten diffusion and (2) use of a filament of the same isotope and composition as that of the crude metal to be purified to eliminate all tungsten from the system.

By increasing the tungsten filament temperature to about 200°C above that necessary to rapidly decompose the metal iodide, thermal cracking apparently occurs prematurely in the vicinity of the filament rather than directly on its surface (Fig. 5), resulting in the requisite annular deposit. This procedure was found effective for producing material that could be easily separated from the tungsten in small pieces.

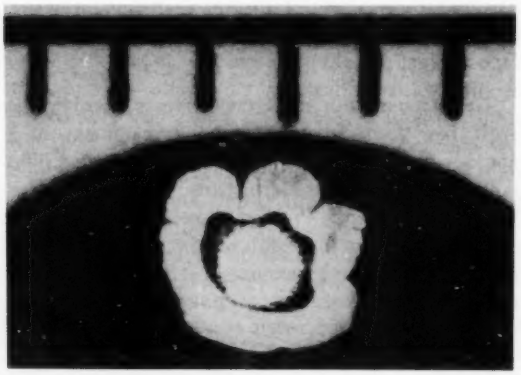

Fig. 5 Annular deposit of zirconium on tungsten showing superficial connection between deposited metal and the filament.

From the purified material obtained by the annular deposition technique, it is usually possible to obtain a single piece of metal which can be extruded into wire a few inches in length. This wire can then be substituted for the tungsten filament and a crystal bar grown using the wire as the nucleus. Resulting bars of metal are then homogenized and further purified by zone refining under high vacuum. Once this final bar is obtained, similar tungsten-free filaments can be fabricated easily, and additional bars can be obtained by repeating the procedure.

Bars of ^{90}Zr, ^{92}Zr, and ^{94}Zr were prepared in this manner for study of the dependence of electrical inversion temperature upon isotopic composition. In this case it was found that a 24-hr anneal (at 800°C) of the zone-melted bars was required to obtain a sharp transition from normal to superconductivity. The metal bars (1.5 in. long, 0.2 in. in diameter) were found to contain no more than three macrocrystals (as twins) in each bar. Similar bars of even-mass titanium isotopes are now being prepared.

The van Arkel–de Boer process has been used to purify Ti, Zr, Hf, Cr, and Nb metals and their isotopes. Purities of >99.9% are almost always attainable, with the major impurity component being tungsten (from the filament). When crystal bars are grown on filaments prepared from the same isotope, purities of 99.99+% are easily obtained. Most metals exhibit enhanced ductility after processing in this manner and have been cold rolled into foils with areal densities of <500 $\mu g/cm^2$; chromium is the single exception. Crystal bars of several metals are shown in Fig. 6.

Vacuum-Evaporation Technology

Vacuum evaporation is a versatile technique for preparing self-supported films of elements of almost any thickness up to 50,000 A and supported films on almost any substrate material. Each element as well as each film thickness poses a different set of preparative problems in vacuum-evaporation technology. Techniques for measurement of film thickness or uniformity of film thickness must also be included as an integral part of any development effort. Since this is a summary report, details on each film preparation are not included, but the results of these developments will be discussed. For general information, a listing of routinely prepared samples is given in the Appendix.

Self-Supported Films

In the past few years, improvements in vacuum-evaporation techniques and vapor condensation methods have permitted a wider range of film thicknesses and sizes. Generally, films of the more reactive metals (e.g., rare earths) have been prepared in conjunction with the reduction–distillation process described earlier.

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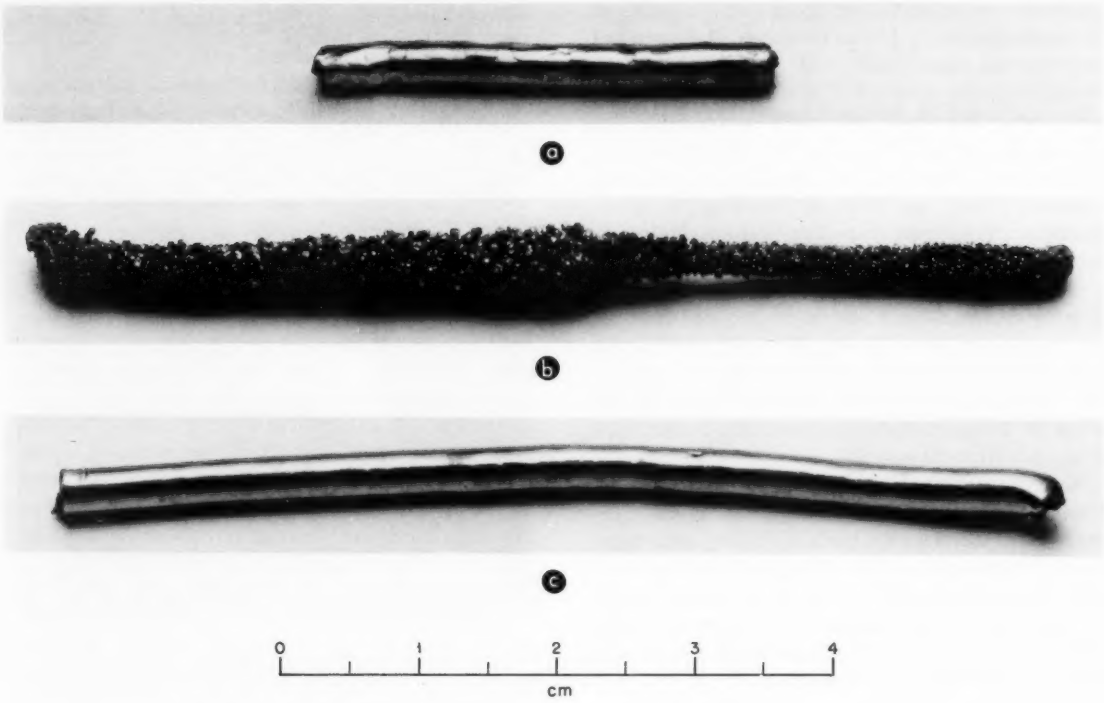

Fig. 6 Crystal bars of (a) zirconium, (b) niobium, and (c) titanium. Note large crystal faces in each bar; these may be used as seeds for single-crystal growing.

Bismuth. Preparation of accelerator targets from elemental bismuth, with self-supported areas of >13 cm^2 and high areal densities, was successful only when vacuum-evaporation techniques were used. Since normal bismuth is mononuclidic and relatively inexpensive, evaporation–condensation could be carried out at large vapor source-to-substrate distances to achieve high uniformity. With a detergent as the parting agent (i.e., a soluble interlayer), 30-cm^2 self-supported condensed films were prepared with areal densities between 3 and 12 mg/cm^2 simply by evaporating the element from a tantalum boat by resistance heating. These films do not float on water since their weight exceeds surface tension. If, however, the substrate and film are soaked in water, the physical strength of the bismuth is sufficient to withstand the tension required to slip the film from the substrate onto a mounting frame of suitable dimensions. Thinner films having areal densities of <1 mg/cm^2 could be stripped from their substrates by flotation on water. A uniformity of ±5% was attainable without difficulty, and only a few pinholes were observed.

Germanium. For more than 2 years, considerable effort has been devoted to the development of a reproducible method for preparing self-supported films of elemental germanium having areal densities between 50 and 1000 $\mu g/cm^2$. Problems involved attainment of sufficient film ductility for nondestructive handling (particularly when areal densities >500 $\mu g/cm^2$ were required) and elimination of pinholes in the films. When germanium vapors were condensed onto a variety of substrates, using a vapor-deposited layer of NaCl as the parting agent, resulting films cracked radially and generally fragmented on being stripped from the substrate. Substrates of many metals and glasses were used with a variety of parting agents without significant success. Semipolished tungsten was the most satisfactory sheet. By taking precautions to cool the substrate and to minimize radiant heat transfer between the vapor source and the substrate by use of multiple heat shields, films of good ductility were obtained. Vapor-condensed NaCl was used as the parting agent. By using 250 mg of germanium metal, reproducible films having areal densities between 950

and 1000 $\mu g/cm^2$ were produced over an area of 40 cm^2. The degree of ductility and structural strength at this thickness permitted mounting of self-supported films over a 1-in.-diameter area.

Manganese. Preparation of high-areal-density, self-supported foils of manganese (>2 mg/cm^2) required considerable development. Manganese in the alpha form (<700°C) is extremely hard and brittle and fractures easily during mounting. This problem was circumvented by forming a vapor-condensed layer on chilled tungsten sheet (polished) coated with a vapor-deposited layer of NaCl (~1 to 2 $\mu g/cm^2$). By this technique, foils with areal densities up to 9 mg/cm^2 (the greatest areal density attempted) were stripped, without fracture, by the technique described for bismuth; the maximum area of product samples was 6 cm^2.

Silicon. Efforts to prepare silicon films of 100 to 2000 $\mu g/cm^2$ areal density by the reduction–distillation method failed because of the high oxygen concentration remaining in the silicon films. Tantalum metal had been used as the reductant, and the high volatility of the SiO species probably resulted in the observed oxygen contamination. However, evaporation of silicon from graphite tubular crucibles using electron-bombardment heating[2] did result in successful preparations. Compatibility of the silicon with the graphite was poor, but rapid evaporation caused only a small loss of isotopes by carbide formation. Silicon vapors were condensed on salt-coated tungsten sheet heated to >350°C. (This minimum temperature was required to obtain suitable epitaxy and to provide films resistant to fracture during stripping and mounting.) Poor results were obtained on cold substrates (amorphous films) because of the flectional weakness of films having an areal density of >100 $\mu g/cm^2$.

Supported Films

Highly adherent coatings of isotopes or normal materials on a variety of substrates have been developed for special projects during the past few years. Only a few of these requiring development of special technology will be discussed.

Magnetically Oriented Iron Films. The preparation of iron films having both specified thickness and magnetic orientation was studied in order to produce specimens suitable for experimental investigation of spin polarization of electrons at the Fermi surface in ferromagnetic materials. (Measurement of tunneling resistance between two similar ferromagnetic films was to be performed with the films magnetized in parallel and antiparallel orientation; the value of the ratio of these resistances, other than unity, would be indicative of partial polarization of electrons at the Fermi surface.) Samples were desired having a sandwich structure consisting of a film of iron, a layer of MgF_2 or SiO, a second layer of iron, and, finally, a cover layer of the dielectric over the entire surface.

These samples were fabricated by evaporation–condensation in a vacuum of $\sim 10^{-8}$ torr using only high-purity (>99.9%) materials. To ensure mobility of the condensing molecules of iron or of the dielectric, the Pyrex glass substrate was heated to 300°C during deposition. All evaporations were performed during a single closure of the vacuum chamber so that exposure of intermediate films to air might be avoided until the final overcoating of dielectric material was complete. To achieve magnetic orientation of the iron films, fixed Alnico magnets were placed on each side of the glass substrate to generate a field of ~20 gauss parallel to the plane of the substrate during deposition. Having the substrate temperature at 300°C enhanced adhesion of the films to the substrate and provided appropriate annealing conditions for the iron deposits.

The ion-pumped evaporator system used to perform these various operations had a pumping speed in the evaporation chamber of ~20,000 liters/sec and was completely bakeable at 450°C. This system was characterized by a very low hydrocarbon contamination in the evaporation chamber compared with other systems utilizing oil-diffusion pumps and cryotrapping. As shown in Fig. 7, electron-bombardment heating devices were employed to evaporate all components of the multilayer sample. Iron was evaporated from a pressed pellet of high-purity powder by driving electrons from a ring filament, immediately above the pellet, into the evaporant; this arrangement achieved volatilization in nearly 2π geometry. By locating the substrate 8 in. above this wide-angle vapor source, excellent uniformity of deposition was achieved over the desired 2-cm-diameter spot.

Multiple masking of the substrate was necessary so that each iron deposit overlapped only one of the gold electrical contacts (formed by vapor condensation in vacuum) and the dielectric deposits completely covered each iron layer. A rotary mask, having three openings, could be indexed over the substrate by manipulation of an ultrahigh-vacuum, rotary feed-through located on the top of the vacuum chamber. The substrate was rigidly mounted on a surface heated by a tungsten filament. Near the bottom of the assembly a quartz

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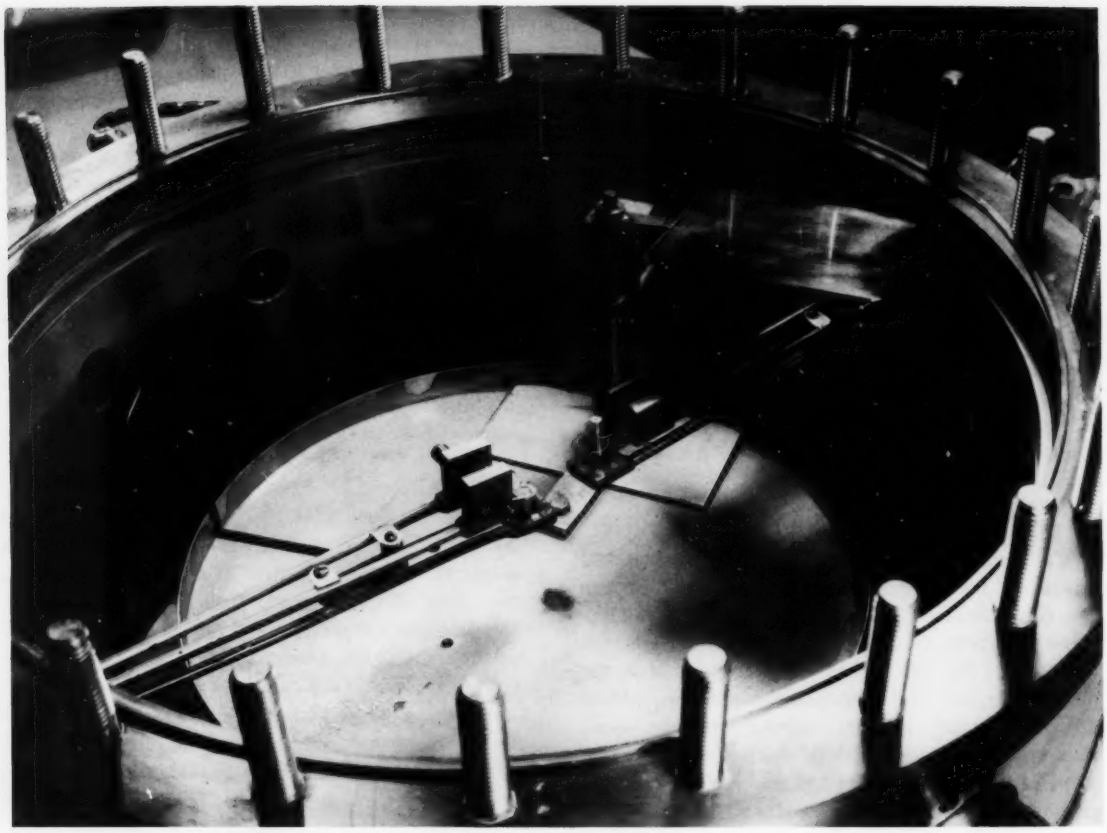

Fig. 7 Electron-bombardment evaporation sources. Left-hand source uses direct electron bombardment of an iron pellet which then serves as its own crucible. Right-hand source uses a tubular crucible and effusion port for evaporation of the dielectric material.

crystal resonator was attached adjacent to the substrate so that the mass of material being deposited during any evaporation could be monitored accurately by measuring the frequency shift of the resonator with reference to a fixed-frequency oscillator. Because of the wide evaporation angle and the distance between the source and the substrate, no significant difference, as determined by weight measurement, was detected between the thickness of deposit as measured by the quartz resonator and that on the glass substrate.

Thickness measurements using the resonating quartz crystals were found to have some temperature dependence. After suitable calibration, however, the estimated error of measurement of mass was found to be ~3%. Temperature variations were minimized by housing the resonating crystal in a metal shell so that only a spot of small, known area was exposed to the vapor beam. Even with this precaution, part of the 3% measurement error could be attributed to temperature variation. A finished sample of the sandwiched films is shown in Fig. 8; the iron layer, deposited directly on the glass substrate, had a thickness of 800 A, the separating layer of SiO about 100 A, and the second iron layer 1600 A. The entire sample was overcoated with >200 A of SiO. Eight similar samples have been prepared in a variety of layer thicknesses.

Thin-Film Platinum Resistance Telethermometry Devices. Measurement of thermal transients created on aerodynamic surfaces by movement of shock waves across the form requires the use of precision fast-response devices whose contour and mounting do not affect the surface smoothness of the device being tested. Telemetry devices such as thermocouples or

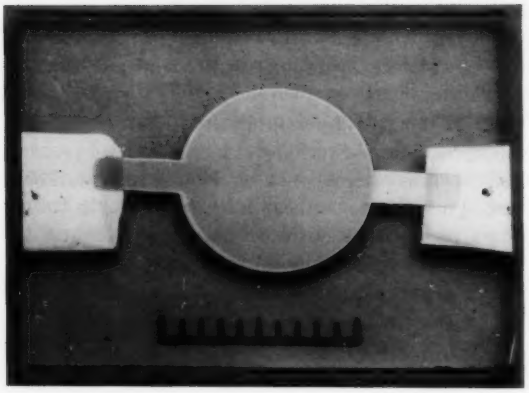

Fig. 8 Magnetically oriented deposits of iron separated by dielectric films on a quartz substrate. This sample was used for tunneling experiments in solid-state physics.

thermistors have thermal response times which are too long for this purpose.

To produce a millisecond-response device, 0.250-in.-diameter quartz disks were employed as substrates onto which a vapor-condensed strip of high-purity platinum was formed (resistance of 100 ± 20 ohms). With a continuous and uniform film, nearly linear response to thermal transients could be observed because of the dependence of the resistance of platinum on temperature. However, pinholes in the film or nonuniformity of the platinum strip could cause failure of the device and nonlinearity of response. Impurities in the film were found to be equally detrimental.

Assurance of high-purity platinum deposits was made by electron-bombardment heating of platinum metal mounted on a tungsten wire so that the platinum created its own "crucible." A heated substrate holder was machined to accommodate 12 quartz disks. A slotted mask through which the platinum vapors passed could be indexed over the holder. Use of 30°-tapered edges on each slot in the mask permitted deposits to be formed with nearly uniform thickness to the edge of the strip. A substrate-to-vapor source distance of 5 in. was needed to attain ±2.5% deposit uniformity. Pinholes in the films were avoided by premelting the platinum in high vacuum and by establishing a uniform metal-evaporation rate before opening a shutter masking the substrate holder. Maximum platinum-to-quartz adhesion was attained at substrate temperatures of >400°C.

After platinum deposition, each disk was overcoated with a dielectric film having an areal density of ~100 $\mu g/cm^2$. Silicon monoxide was found to be more satisfactory than MgF_2 or SiO_2 for this purpose. Before dielectric deposition, silver metal electrical contacts were painted onto the sides of each disk and were extended to the back face for later welding of 0.002-in.-diameter wire connectors. Each disk was heated in air to 400°C to cure these painted surfaces. As the final step, the SiO film was deposited over the entire surface. Figure 9 illustrates several of the finished telethermometry devices.

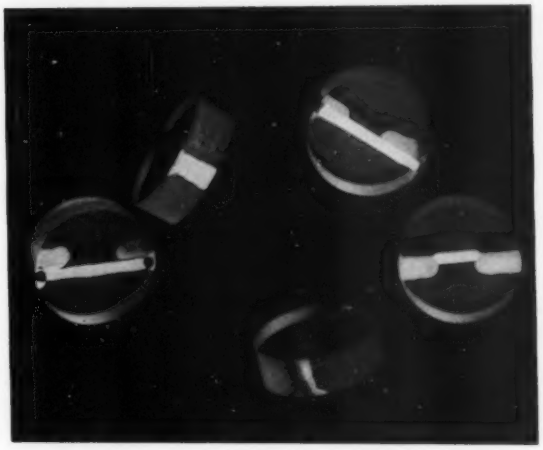

Fig. 9 Telethermometry devices formed by vacuum evaporation–condensation of platinum onto $\frac{1}{8}$-in.-diameter quartz substrates.

Tellurium Coatings on Stainless Steel and Other Substrate Materials. Tellurium is generally considered to be a nonmetal and has a high vapor pressure (0.1 torr at 433°C). Since the adherence of metal coatings on surfaces depends largely on the amount of diffusion bonding or chemical interaction between the coating material and the substrate, vapor-deposited layers usually are most adherent if formed at substrate temperatures in excess of 350°C. With tellurium, such substrate temperatures would be prohibitive since the "sticking" coefficient of tellurium would be much less than unity because of its high vapor pressure. Thus development of techniques applicable to the formation of adherent tellurium coatings was necessary.

Diffusion of tellurium into stainless steel or glass at low temperatures (<100°C) was found to be too slow for good adherence of vacuum evaporated–condensed

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films; coatings prepared at these temperatures were easily removed by gentle wiping (no abrasion). Formation of a thin layer of an active-metal telluride seemed the most feasible approach to the solution of this problem provided that a suitable active metal could be made to adhere to the desired substrate and that the quantity of this metal (thickness) was insignificant for whatever purpose the finished sample was to be used. Only those metals were considered which could be vapor deposited at sufficiently high substrate temperatures to form adherent chemical or diffusion bonds with the substrate (e.g., one of the rare earths, since rare earths easily form tellurides and tend to form highly adherent coatings on most clean metal surfaces).

Deposits of erbium metal, as little as 50 $\mu g/cm^2$, on stainless steel or glass were suitable for forming adherent deposits of tellurium having areal densities up to 2 mg/cm^2. As might be anticipated, however, this multilayered coating was similar to a graded glass-to-metal seal, i.e., the gross properties of the tellurium deposit were those of the elemental tellurium, and not of the minute amount of erbium telluride formed at the interface. For samples to be employed at temperatures $<200^\circ C$, the tellurium deposits were found to be adherent, hard, and durable, and the abrasion resistance proved to be similar to that of the bulk material. However, the tellurium coatings were easily removed (for all practical purposes) in vacuum by heating the substrate to $>200^\circ C$.

Application of this technique to the preparation of elemental or oxide deposits of radioisotopes (e.g., the actinides) on incompatible substrates has been successful. Of particular importance has been the use of flash coatings to form adherent bonds between hydrides and metal substrates having widely differing coefficients of expansion or crystal-lattice parameters. (See discussion of the HENRE tritium-target preparation on page 14.) In general, the use of chemical bonding to form adherent coatings of nonmetals on almost any surface has general applicability.

Rolling of Metals

As a general preparative method for isotope targets, mechanical rolling of high-purity metals is the preferred technique whenever feasible, since little or no loss of material is experienced in forming films by this method. Rolled foils have physical properties identical to those exhibited by bulk material, with the exception of microscopic crystallite distortion and work-induced strains. Both of these physical deviations can usually be removed by appropriate annealing of the foil. The material strength and uniformity of thickness (when compared with evaporated–condensed films) which can be obtained by rolling small quantities of metals are advantageous.

Over the past 3 years, numerous improvements in rolling facilities and techniques have permitted preparation of foils thin enough to be transparent. These developments have not been restricted to stable-isotope target preparations but have been used with marked success in forming thin foils of many of the heavy elements, particularly uranium and plutonium.

Stable-Isotope Foils

High-purity metal preparation by the simultaneous reduction–distillation technique has made many metals available with the required qualities for cold rolling into thin foils. Application of the van Arkel–de Boer iodide process for obtaining tungsten-free metallic titanium and zirconium isotopes has also permitted fabrication of larger and thicker samples of these metals by cold rolling.

Metals produced by the reduction–distillation process (lanthanum or thorium reductant) have improved hardness and ductility compared with commercial materials. The rare-earth metals (excluding cerium and lanthanum) have been cold rolled into foils of $>6\text{-}cm^2$ area with a minimum areal density of ~ 0.8 mg/cm^2 (3×10^{-5} in. thick), and most could be reduced to areal densities of 0.3 to 0.5 mg/cm^2, provided that the sample size was restricted to ~ 1 cm^2. Reactivity of these metals with oxygen and nitrogen is greater than for bulk materials, and all rolling to thicknesses <0.005 in. was performed in a purified argon atmosphere containing no more than 5 ppm of these gases.

Similarly the reactive Group II-A metals were prepared in thin-foil form. Barium isotope metal (reduction–distillation produced) was rolled into self-supported foils of $\sim 2\text{-}cm^2$ area with an areal density of 0.5 mg/cm^2. These foils were transparent to strong light. Magnesium metal, being somewhat harder than heavier elements in the same group, did not adhere to the rollers and was rolled into foils having a minimum areal density of 0.1 mg/cm^2 (2×10^{-5} in. thick).

The minimum areal densities attainable for these various metals by cold-rolling techniques are approaching (and in some cases are well within range of) the thicknesses required for target foils used in charged-particle interaction studies (see Appendix). In many cases the gap which existed only a few years ago between the maximum thickness of self-supported

targets obtainable by vacuum evaporation–condensation techniques and the minimum thickness to which the same metal could be cold rolled has disappeared. Future development efforts will be directed toward the design, construction, and application of a miniature "cluster" mill which has small-diameter work rolls and is compatible with vacuum–inert gas environments. With such a mill, it is expected that foils of larger area will be prepared in the areal density range 0.1 to 0.5 mg/cm^2 for most pure metals.

Radioisotope Foils

As with stable-isotope metals, the physical properties of metallic radioisotopes are related to the purity of the metal. Inclusion of oxides, nitrides, or fluorides, and of some metallic contaminants, causes embrittlement which precludes preparation of thin self-supported foils by cold rolling. Because of limited processing facilities, especially shielded enclosures, all work in radioisotope metal-foil preparation has been confined to alpha-emitting isotopes which can be obtained in high chemical purity; of these, only uranium and plutonium isotopes will be discussed.

In cooperation with the Center, the Development Division (Y-12 Plant, Oak Ridge) and Rocky Flats (Dow Chemical Company) personnel developed chemical conversion and reduction techniques for uranium and plutonium isotopes which permit as little as 5 g of oxide to be processed into metallic form suitable for cold rolling. The process converts UO_2 and PuO_2 to the fluorides and, through a subsequent bomb reduction with calcium and iodine, produces consolidated metal. Although this process is quite similar to that used in the large-scale production of these metals, many improvements were necessary to permit 5 g of material to be produced with 75 to 90% yield and attainment of a high-purity product compatible with preparation of thin foils. The details of the plutonium process can be found in a published report by Conner;[4] the work on uranium is not yet published.

Over the past year many samples of uranium and plutonium isotopes were rolled into thin foils. Until this facility existed, the thinnest reported rolled uranium foil was ~5 × 10^{-4} in., and plutonium had not been rolled to below 1 × 10^{-3} in. The lower thickness limit attained by Center personnel on both of these metals is 4 × 10^{-5} in. (i.e., an areal density of ~2 mg/cm^2). Because of the high density of these metals, the foils are not transparent to light at this thickness level. The reactivity of both elements is extreme at thicknesses below 2 × 10^{-3} in. Spontaneous ignition can occur on exposure to the atmosphere, and all rolled samples are packaged in sealed glass ampules under vacuum or in a purified argon atmosphere.

As with other thin foils, the maximum area of a rolled foil, for a given amount of starting material, is dependent on foil thickness. However, pure uranium and plutonium isotopes have excellent flectional and ductile properties, and foils of >6 cm^2 have been prepared even at the low thickness limit. Several circular foils of 99+% ^{235}U metal were prepared having a thickness of 2 × 10^{-4} in. and an area of 125 cm^2; these samples were for use in a fission chamber. For the first time, metal foils of isotopically enriched ^{236}U (99.8%) and ^{241}Pu (99.9%) were prepared; foils of ^{236}U were rolled to a thickness of 1 × 10^{-4} in. and a circular area of 13 cm^2, and ^{241}Pu metal was prepared in rectangular foils of 12 cm^2 with a thickness of 5 × 10^{-4} in.

Radioisotope Targets and Sources

Radioisotope Facility

A new radioisotope containment area for the preparation of targets and sources was constructed and equipped. In designing this facility, emphasis was placed on handling heavy elements, principally the actinides and other alpha-emitting materials. Otherwise, only weak beta- and/or gamma-emitting isotopes can be accommodated because of minimal shielding provided on the containment glove boxes surrounding fabrication equipment. Preparative techniques in this area are limited to vacuum evaporation–condensation, rolling, tritium sorption, and some electrodeposition; Tungsten–Inert Gas (TIG) welding equipment for source encapsulation is also included.

Two rolling mills in this area are enclosed in glove boxes in which purified argon atmospheres can be maintained. One mill is an 8-in., 4-high/2-high convertible Stanat machine capable of exerting a 50-ton pressure on the rolls, while the other is a 2-in., 2-high chain-driven mill suitable only for relatively soft materials in small quantities. Argon-atmosphere annealing furnaces are enclosed within the glove box of the Stanat mill.

Steel glove-box enclosures for the various vacuum-evaporation systems are designed to handle only one element (and/or its isotopes) in order to limit cross contamination between the different alpha- and beta-emitting heavy-element isotopes. To date, contamination has never exceeded 0.01%. At present, evaporation

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facilities are available for Th, U, Pu, Np, Am, Cm, and Cf isotopes.

Californium-252 electrodeposition and/or molecular plating equipment is enclosed in a single isolated containment zone. This system can handle 10 μg of californium when necessary, but usually <5 μg is used at any one time. No other heavy elements are formed into targets or sources by electrodeposition since vacuum-evaporation methods have proved to be superior for this purpose.

Two nonconnecting, isolated glove boxes have been equipped with vacuum-evaporation equipment for forming special sources of ^{210}Po. Because of the special handling necessary with this isotope in elemental form, these systems could not be included in the complex previously described.

Each vacuum-evaporation system used for heavy-element target and source preparation is of the oil (DC-704)-diffusion type which has an isolating high-vacuum valve between the chamber and the 4-in. pump. Rough pumping of the systems is performed with a 1397-B Welch mechanical pump separately enclosed in a glove box. All exhaust gases from the pumps and glove boxes are vented to the cell ventilation system through absolute filters (0.3 μ); air intake to the containment enclosures is also filtered, and each box is maintained at a minimum negative pressure of 0.2 in. water. Other than arrangements for backfilling the vacuum chambers (when necessary) with argon, no provision is provided for maintaining an argon atmosphere in the evaporation boxes. Each system is equipped with appropriate evaporation sources of the electron-bombardment and resistance-heating types.

Also included within the target facility are the various systems for preparation of tritium-containing targets of all sizes from <1.25 up to 22 in. in diameter. Two vacuum-evaporation systems and two tritium-sorption systems (glove boxed) are integral parts of this operation.

Considerable emphasis has been placed on quality control of radioisotope targets, since the chemical purity of these materials is generally less than that of stable isotopes. A variety of counting devices and other measuring instruments have been included. Of particular importance are two low-geometry counting chambers using solid-state detectors where count rates can be obtained from fabricated samples and semiquantitative identification of radioisotopic impurities can be made. Ion chambers have also been provided for high-level samples such as ^{210}Po and ^{242}Cm deposits. Alpha, beta, and fission-fragment counting can all be performed on an absolute basis.

Targets and Sources Produced by Vacuum-Evaporation Methods

More than 300 targets and sources have been prepared in the radioisotope facility by vacuum evaporation–condensation since operation began in March 1967. Many of these samples were of the self-supported variety, produced by evaporation of oxides or fluorides of the heavy-element isotopes on thin carbon substrates (substrate areal density 10 to 50 μg/cm^2). Most samples, however, were deposited films or coatings on heavy substrate materials having a thickness of 1×10^{-3} in. or more. Isotopes of Pb, Th, U, Np, Pu, Am, Cm, and Cf have been deposited in both self-supported and supported forms. Since each target preparation involves a variety of routine operations described elsewhere,[2] only a few unusual preparations will be discussed.

Fission-Chamber Plates of PuO_2 on Aluminum. Accurate measurements of the fission cross section of ^{239}Pu are being performed by the Neutron Physics Division of ORNL using a fission chamber containing 21 plates of 5×10^{-3}-in.-thick aluminum with a diameter of 3 in. and coated with high-assay ^{239}Pu. Nineteen plates were coated on both sides with PuO_2 by evaporation–condensation to an areal density of 1 mg/cm^2; the two terminal plates of the fission chamber were coated only on one side.

The specified stacking of the plates in use required that the coated substrates be planar (no significant warpage). Vacuum annealing of the substrates between planar copper blocks at 450°C proved to be satisfactory, provided that a minimum temperature of 350°C was maintained on the substrates during PuO_2 vapor condensation (to ensure good film adherence) and that the coated disk never exceeded the 450°C annealing temperature.

By using electron-bombardment heating of a tungsten cylindrical crucible (1.25 in. long, 0.20 in. inside diameter), evaporation of the oxide at ~2400°C was performed without difficulty. With appropriate evaporation geometry, the substrate rotation over the vapor source resulted in uniform deposition. About 15 g of the oxide was required to coat all disks.

Curium-244 Cross-Section Samples. Samples of ^{244}Cm were required for fission cross-section measurements in which the atom count of the curium was known to an accuracy of >95%. A vacuum-deposited curium oxide layer of ~50 μg/cm^2 areal density over a circular spot of 0.25 in. diameter on a 0.423-in.-diameter polished platinum disk was required.

However, owing to the fact that the available curium oxide was impure and the true nature of a vacuum evaporated–condensed deposit is difficult to define without extensive chemical and/or spectrographic analyses, initial attempts to produce a variety of samples ranging in areal density from 10 to 100 $\mu g/cm^2$ (determined by direct weighing of the deposit) were unsuccessful. Subsequent alpha counting of these samples at Los Alamos Scientific Laboratory (LASL) proved that the quantity of curium present was less than expected by a factor of 200.

Low geometry (i.e., known source-to-aperture distance; generally used for high-level counting) alpha-counting equipment was designed to resolve this difficulty. An 87-cm distance between the radioisotope source and a silicon diffused-barrier detector was used, and the counter system, together with a 256-channel analyzer and other electronic components, was able to resolve the two alpha-particle emission energies (5.80 and 5.76 Mev) of the ^{244}Cm species. The amount of curium present was then determined to a calculated accuracy of ±0.5%. It was found, however, that for curium targets of different thicknesses the quantities of curium deposited, as determined by low-geometry counting, were less than those determined by direct weight measurement. Deviation factors of between 10 and 200 were observed, but with no apparent systematic variation. To obtain reproducible targets having the desired ^{244}Cm concentration, fractionation during vacuum evaporation was necessary. During initial heating of the evaporant in a tungsten tubular crucible, all vapors emitted before reaching the evaporation temperature of the CmO_2—i.e., 2000°C—were condensed on a shield separating the vapor source and the target substrate. When the crucible finally reached 2000°C, the condensing shield was removed, and evaporation–condensation of the residual material was allowed to proceed. Variations between mass measurements and ^{244}Cm assay by counting were found to be at least reasonably constant by this procedure; the difference factor was usually reduced to <10.

Three separate ^{244}Cm targets were prepared having a nominal areal density of 55 ± 1 $\mu g/cm^2$. Counting data agreed with experimental data obtained at LASL to within ±1%. It was concluded that weight measurements of vapor-deposited radioisotope targets (heavy-element materials) could not be relied upon to produce accurate atom-count information and only direct counting techniques would provide satisfactory analytical data unless pure starting materials of known composition were used.

Polonium-210 Coatings on Aerodynamic Forms. A process was developed to form adherent deposits of elemental ^{210}Po, and several special forms were coated using this technique. Polonium exhibits difficulties in forming adherent coatings because of its high vapor pressure at low temperatures. Since the forms were to be used to study the effects of ionization for high-velocity and static gas viscosities, good adherence of the coating was mandatory to create a constant alpha-emission intensity (the ionization source) and a low radioactive contamination potential, particularly at high gas velocities. As with tellurium, erbium metal subcoatings were employed to chemically bond the polonium to the substrates.

With polonium the additional problem of recoil sputtering was encountered, which resulted from the high-energy (5.30 Mev) alpha emission. Material loss by this mechanism was reduced by overcoating the polonium film with a sufficiently thin (areal density ~100 $\mu g/cm^2$) metallic chromium coating to permit nearly all the alpha particles to penetrate without significant energy attenuation. The thickness of the combined polonium coating film and the chromium overlayer was designed to permit maximum ionizing radiation (~35 mc/cm^2) to enter the gas phase surrounding the source without losing appreciable polonium.

The special forms coated were a 3-in.-diameter aluminum disk (0.060 in. thick) and a 3.5-in.-long 9° right circular cone made by machining pure titanium bar stock. In both cases the surfaces were precoated with chromium to form an adherent bond between the aluminum or titanium substrate and the erbium metal layer. Each metal film, including the ^{210}Po, was deposited by vacuum evaporation–condensation techniques. Chromium and erbium metals were evaporated by electron-bombardment heating of a pellet of each of these materials, while polonium was evaporated from a tantalum tubular crucible heated by electrical resistance. Polonium electrodeposited on platinum gauze was used directly as the evaporation charge.

The most difficult problem was holding to the maximum atom density deviation of ±5% for the polonium deposit on the conical surface. Obtaining uniform films of chromium and erbium was straightforward since natural materials were being used and vapor source-to-substrate distances could be large. Only three times the required quantity of ^{210}Po was available; therefore special evaporation geometries were studied and tested, using tellurium as the evaporant, before deposition of polonium was attempted. By calculation, a deviation in film thickness of ±7.5% from

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the mean value over the entire conical surface could be attained by using two tubular crucible evaporation sources whose axes were perpendicular to the surface of the cone and which were located at the ends of the cone. However, testing of the system was required to determine the correct distance between the vapor sources and the conical surface.

The conical titanium substrates are illustrated in Fig. 10 with a segmented solid cone used to determine (by weight measurement) the areal density of tellurium films deposited at various distances from the vapor source. During evaporation the cones were rotated at ~100 rpm in such a position that the surface was always parallel to a line connecting the two vapor sources.

Fig. 10 Right-circular titanium cones (9°) to be used in aerodynamic studies. These cones were coated with chromium and ^{210}Po. The segmented cone was used to determine coating uniformity and evaporation geometry by direct mass measurement.

The alpha emission from one cone (coated with the various materials but prior to application of the chromium overcoating) was 18 mc/cm^2. The chromium overcoating was found to limit the dispersion of contamination to acceptable levels even when the sample was subjected to air velocities approaching Mach 3. The most active specimen of the several disk samples had an alpha emission from the surface of 38 mc/cm^2. As with the conical sample, only minor contamination was observed after using the disk in an oscillating gas viscometer for 14 days.

Special Projects

Targets for the High Energy Neutron Reaction Experiment (HENRE)

As previously reported,[1] Project HENRE required the production of a neutron field of ~10^{13} n/sec from the 1000 cm^2 surface for four consecutive hours. By utilizing the ^{3}H(d,n)^{4}He reaction, such a field was considered feasible, provided a tritium-containing target could be fabricated which could withstand an estimated continuous deuteron beam power of 75 kw without massive deterioration. To produce such a target, a water-cooled copper substrate was designed having a 1000-cm^2 surface on which could be deposited a layer of erbium tritide. This tritium-containing compound was chosen because of its high thermodynamic stability and resistance to decomposition under deuteron bombardment.

The active target surface was formed by vacuum evaporation–condensation of several metal films onto the copper substrate. A graded seal was developed to gain maximum adherence of the erbium to the substrate and thus provide the best heat-transfer properties. Each copper substrate was first coated with a film of chromium metal to an areal density of ~250 μg/cm^2 while the substrate was heated to ~350°C; onto this surface erbium metal was condensed in a layer having an areal density of 2 to 2.5 mg/cm^2. This composite structure was able to withstand repeated thermal cycling from room temperature to 450°C and formed an adherent layer capable of maintaining its integrity under bombardment at current densities of >75 watts/cm^2, even after sorption of tritium into the erbium film.

The vacuum environment and method of deposition of the various metal coatings were most closely related to the neutron-generating capability of the finished product. The required vacuum levels of 10^{-7} to 10^{-8} torr were achieved during evaporation–condensation by a 500-liter/sec ion pump and a titanium sublimation pump, the combined capacity of which was estimated to be 20,000 liters/sec. To gain maximum uniformity of the vapor-deposited layers, the target substrate was rotated at 100 rpm behind a mask so that metal vapors were condensed over a semicircular area equal to half the total target area at any instant. By maintaining a substrate-to-vapor source distance of 15 in., a uniformity in areal density of 93% was achieved over the entire 1000-cm^2 target area.

Preparation of one such target required the evaporation of ~7g of chromium metal, ~65 g of

erbium metal while maintaining the rotating substrate at >350°C, and a total of 6 hr, not including the time consumed in reaching the appropriate substrate temperature and in cooling after evaporation–condensation was complete. Electron bombardment was used exclusively for heating cylindrical bars of the various metals. A thin coating of cadmium metal (vacuum evaporated from a resistance-heated source) was deposited over the active target surface after it had cooled. This coating was used to inhibit surface oxidation of the erbium film during transfer of the target to the tritium-sorption system.

Tritium sorption was performed in an ion-pumped, all-metal system (see Figs. 11 and 12), where the substrate was heated to ~450°C under ultrahigh vacuum; the pressure attainable in the chamber was measured at 2×10^{-9} torr. The tritium gas was purified by passing it over hot uranium chips and then sorbing it on cold uranium as uranium tritide. Three uranium traps were used simultaneously to maintain a maximum inventory of 250,000 curies of tritium gas.

As can be seen in Fig. 12, the target substrate was placed on a metal ring immediately above a tungsten wire heater. This configuration permitted heating the entire substrate to any desired temperature up to 475°C. During the heating process the cadmium overcoating was removed by evaporation and was condensed on the system walls, thus permitting contact between the tritium gas and the uncovered erbium-metal sorption surface. When the target reached its maximum temperature and pressure equilibrium had been established (10^{-8} torr), the ion pump and titanium sublimator were isolated, and tritium gas was admitted to the system. Pressures of tritium between 180 and 450 torrs were used. After a "soak" period of 1 hr at the highest target temperature, the system was cooled to room temperature, the excess tritium gas was readsorbed on the uranium in the storage traps, and the

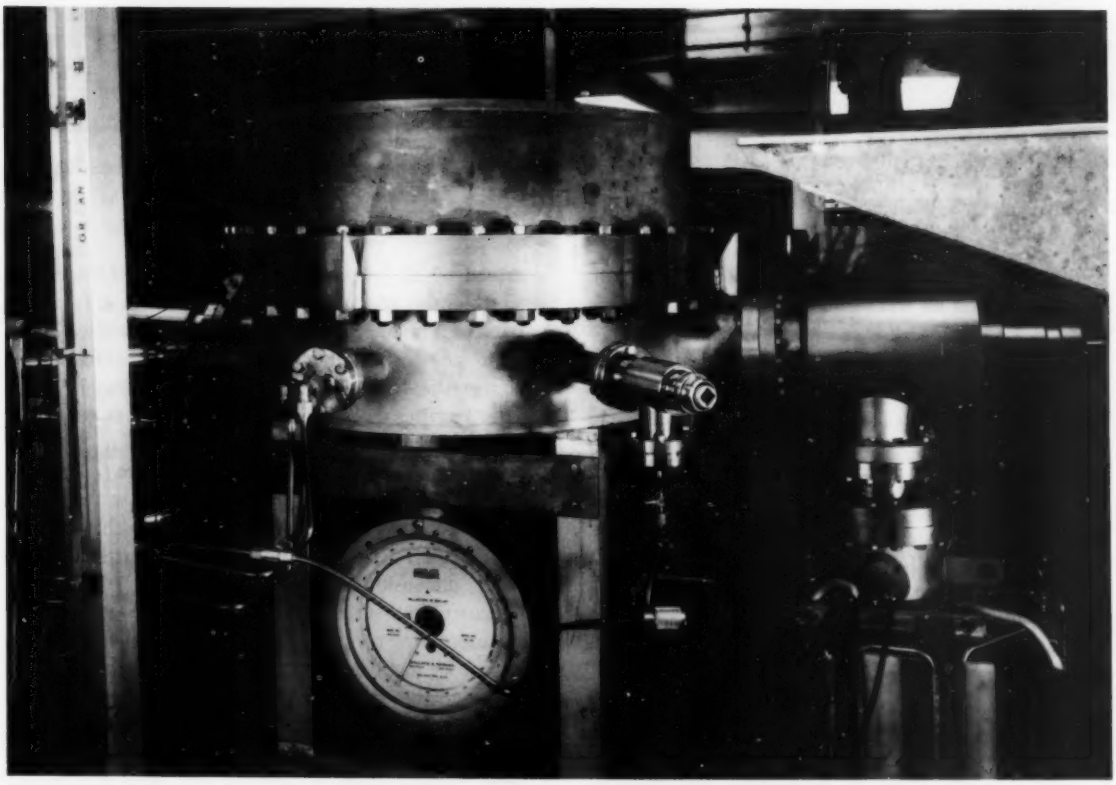

Fig. 11 Tritium-sorption system for preparation of HENRE targets. Entire system is stainless steel, has metal-to-metal seals, and is capable of being evacuated to 2×10^{-9} torr. The 200-liter/sec ion pump is shown in the lower right-hand corner.

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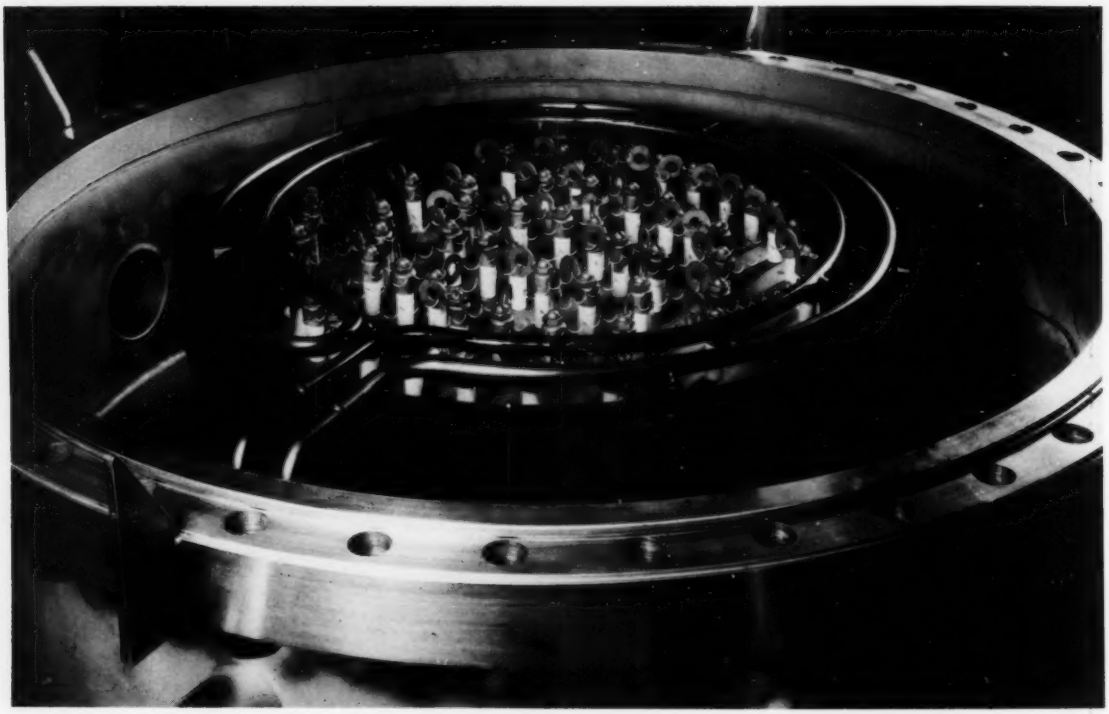

Fig. 12 Heater assembly interior to the tritium-sorption system. The 40-lb copper substrate rests on the rings immediately above the coiled tungsten heater.

system was brought to atmospheric pressure with argon. A plastic lid was bolted over the target surface (not in contact with the erbium tritide layer) to preserve an argon atmosphere until the target could be used.

Surfaces deposited on the target by this fabrication technique were usually of matte appearance and produced a bremsstrahlung level of 10 rems at 6 to 10 in. from the surface. Under deuteron bombardment these targets generated a field of 5×10^{12} to 3×10^{13} n/sec for at least 4 hr (nonpulsed beam); several were bombarded for more than one period, with average yields varying from 10^{12} to 10^{13} n/sec. Forty such targets were prepared and at least 20 more are to be fabricated during the next year.

Preparation of High-Purity Calcium Disks

One hundred disks of high-purity normal-assay calcium metal with a diameter of 2 ± 0.001 in. and a thickness of 0.010 ± 0.001 in. were prepared. High-quality 1-in.-diameter calcium metal bars were obtained which had a chemical purity of 99.9%. Spark-source mass spectrometric analyses of these 6-in.-long bars were made (at the ends and in the midsection of each bar) to obtain a surface-impurity profile of the starting material before, and cross-sectional profile after, cutting the bars into $\frac{1}{8}$-in.-thick disks. Each disk was then rolled into a sheet, using a 3-in. tool-steel rolling mill in a purified argon atmosphere (containing a total of <3 ppm oxygen, nitrogen, and water vapor). The sheets were cut in a precision punch. Measurements of all disks prepared indicated that the samples were dimensionally accurate to $\pm 5 \times 10^{-4}$ in. in diameter and thickness. Spectrometric analyses of representative metal disks indicated that no chemical impurities had been added during this fabrication as far as could be detected (<1 ppm); the oxygen and nitrogen contents of the samples remained unchanged.

All calcium disks and associated scrap material were shipped under vacuum ($<10^{-6}$ torr) in stainless-steel containers that had been prebaked under vacuum. Subsequent testing of this material indicated the metal was suitable for the specified purpose.

Ultrahigh-Vacuum Electron Microscope

A standard Hitachi HU-11-B3, metallurgy-type electron microscope was purchased for use in examining thin isotope films prepared by the Isotope Target Center. This instrument will be used in:

1. Quality control of isotope foils being produced.
2. Research and development of techniques for forming amorphous, polycrystalline, and/or single-crystal thin films.

Commercially available electron microscopes normally operate at 10^{-4} to 10^{-6} torr and are available only with oil-diffusion-pumped columns. Greased O-ring seals for vacuum-atmosphere feed-throughs are used to transmit either rotary or linear motion (or both) from control rods outside the microscope to the various apertures and stages within. Subsequent contamination from hydrocarbon vapors originating in these greases, together with air inleakage through the seals, permits only short-term examination of specimens and makes this type of construction unsuitable for present purposes.

The Center's Hitachi instrument is being modified to permit its operation at pressure levels of $\leqslant 10^{-8}$ torr. All feed-throughs will be replaced with "wobble-pin" bellows seals for transmission of linear motion. If baked and outgassed Viton elastomer O rings and/or copper gaskets are used as static seals, little or no contamination will result, and ultrahigh vacuum can be achieved.

A new pumping system was also designed and constructed for the instrument. Three differential-sputter ion pumps (two with a capacity of 100 liters/sec and one of 20 liters/sec) are used to differentially evacuate the entire microscope column. Each 100-liter/sec ion pump is coupled with a 1000-liter/sec titanium sublimation pump, and all pumps are connected to the column through stainless-steel manifolds (2 or 3 in. in diameter) fitted with stainless-steel bellows to aid in alignment of the components and to dampen vibrations transmitted through the pump support rack. (PSB)

Publication

E. H. Kobisk, Isotope Targets for Nuclear Research, *Nucleonics*, **24**(8): 122-124 (August 1966).

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(The Appendix follows on pages 18 and 19.)

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Appendix: Target Preparations

Legend: a—evaporation; b—rolling; c—electrolytic deposition; d—casting or pressing; e—miscellaneous; 1—self-supporting; 2—metal backing; 3—thin carbon

Element	Range of thickness, μg/cm²	Method of preparation	Backing	Form of deposit
Aluminum	5 to 12,000	a	2	Al
	150 up	a	1	Al
		d	1	Al
Americium	Undetermined	a		Am or oxide
Antimony	500 to 3000	a	1	Sb
Arsenic	10 to 200	a	2	As
Barium	10 to 1000	a	2	Salt
	2000 up	d	1	Salt
		a, b	1, 2	Ba
Beryllium	Undetermined	a	1, 2, 3	Be
Bismuth	500 to 15,000	a	1	Bi
	10 to undetermined	a	2	Bi
Boron	20 to 250	a	1	B
	10 to 1000	a	2	B
Bromine	10 to 1000	a	2	Bromide
	5000 up	d	1	Bromide
Cadmium	10 to 200	a	2	Cd
	1000 up	b	1	Cd
		d	1	Cd
Calcium	10 to 1000	a	2	Ca
	700 up	b	1	Ca
		d	1	Ca or carbonate
Californium	100	c	2	Oxide
Carbon				
Normal	1.5 to 200	a	1	C
Isotopic	10 to 200	e	2	C
Cerium	10 to undetermined	a	2	Ce oxide
Cesium	10 to 1000	a	2	Salt
Chlorine	10 to 1000	a	2	Salt
	5000 up	d	1	Salt
Chromium	500 to 12,000	a	1	Cr
	10 to undetermined	a	2	Cr
		d	1	Cr
Cobalt	1000 up	b	1	Co
	10 up	a	2	Co
	100 to 1000	a	1	Co
Magnesium	150 to 1000	b	1	Mg
	Up to 20,000	a	1	Mg
		d	1	Mg
Manganese	300 to 1600	a	1	Mn
	10 to undetermined	a	2	Mn
Mercury	1000 to 4000	c	on Au	Hg
	Undetermined	a	2	Salt
Molybdenum	10 to 1000	a	2	Mo
	1000 up	b	1	Mo
Neodymium	10 to undetermined	a	2	Nd
	1000 to 15,000	b	1	Nd
Neptunium	10 up	a	2, 3	Oxide
Nickel	500 up	b	1	Ni
	10 up	a	2	Ni
		d	1	Ni
Niobium	10 up	a	2	Nb
	1000 up	b	1	Nb
Palladium	1000 up	b	1	Pd
	10 up	a	2	Pd
		d	1	Pd
Platinum	4000 up	b	1	Pt
	10 up	a	2	Pt
		d	1	Pt
Plutonium	10 up	a	2	Oxide
	5000 up	b	1	Pu (1% Al)
Potassium	7500 to any cast size	b, d	1	K
Promethium	10 up	a	2	Pm oxide
		a	2	Pm
Radium	10 up	a	2	Bromide
Samarium	1000 to undetermined	b	1	Sm
	10 up	a	2	Sm
Scandium	10 up	a	2	Sc
	1000 up	b	1	Sc
Selenium	300 to 5000	a	1	Se
	10 to 5000	a	2	Se
		d	1	Se

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Copper	500 up	b	1	Cu
	10 to 1000	c	2	Cu
	200 up	a	1	Cu
		d	1	Cu
Curium	10 up	a	2, 3	Oxide
Dysprosium	2000 to 25,000	b	1	Dy
	10 up	a	2	Dy
Erbium	10 to undetermined	a	2	Er
	2000 up	b	1	Er
Europium	10 to undetermined	a	2	Eu
	1000 up	b	1	Eu
Fluorine	25 to 500	a	2	Fluoride
Gadolinium	2000 to 25,000	b	1	Gd
	10 up	a	2	Gd
Gallium	10 to undetermined	a	2	Ga
	10 to 200	a	3	Ga_2O_3
		d	1	Ga_2O_3
Germanium	10 up	a	2	Ge
	300 to undetermined	a	1	Ge
Gold	10 up	a	2	Au
	Undetermined	a	1	Au
		d	1	Au
Indium	5000 up	b	1	In
	10 up	a	2	In
		d	1	In
Iridium	10 to 100	a	2	Ir
Iron	500 up	b	1	Fe
	10 up	a	2	Fe
		d	1	Fe
Lanthanum	10 to undetermined	a	2	La
	2000 up	b	1	La
		d	1	La
Lead	8000 up	b	1	Pb
	500 to 8000	a	1	Pb
	10 to 1000	a	2	Pb
		d		Pb
Lithium	4000 to any cast size	b, d	1	Li
	10 to 500	a	3	LiH
	10 to 500	a	2	Li
Lutetium	10 to undetermined	a	2	Lu
Silver	1000 up	b	1	Ag
	100 to 5000	a	1	Ag
	10 to 1000	a	2	Ag
		d	1	Ag
Silicon	10 to 1000	a	2	Si
	50 to 400	a	1	Si
	10 to 150	a	2, 3	SiO_2
Sodium	7500 to any cast size	b, d	1	Na
Strontium	2000 up	b	1	Sr
Sulfur	5000 up	d	1	S
	10 to 100	a	2	S
Tantalum	1000 up	b	1	Ta
	10 to 1000	a	2	Ta
Technetium	10 to 1000	a	2	Tc
Tellurium	10 to 1000	a	2, 3	Te
		d	1	Te
Thallium	8000 up	b	1	Tl
	10 up	a	2	Tl
Thorium	4000 up	b	1	Th
	10 up	a	3	Th
Tin	4000 up	b	1	Sn
	10 to 1000	a	2	Sn
	500 to 4000	a	1	Sn
Titanium	500 up	b	1	Ti
	10 up	a	2	Ti
Tritium*	100 to 5000	e	2	Metal
Tungsten	In Cu matrix	b	2	W
	2000 up	d	1	W
Uranium	10 up	a	2	Oxide
	5000 up	b	1	U
Vanadium	1000 up	b	1	V
	10 up	a	2	V
Ytterbium	10 to undetermined	a	2	Yb
	1000 up	b	1	Yb
Yttrium	10 to undetermined	a	2	Y
	700 up	b	1	Y
Zinc	1000 up	b	1	Zn
	10 up	a	2	Zn
Zirconium	500 up	b	1	Zr
	10 to 5000	a	2	Zr

*Adsorbed on Zr, Y, Ti, or Er.

Miniature ^{125}I X-Ray Source

The availability of a portable ^{125}I X-ray unit (trademark: Söremark X-ray unit) about the size of a pocket pen (Fig. 1) has been announced[1] by the Swedish Atomic Energy Company. The radiation source consists of ^{125}I deposited on a 0.5-mm-diameter wire and is sealed in a metal capsule. Source activities of up to 0.5 curie are offered. During exposure the source is moved to the thin aluminum window in the mounting (Fig. 2), through which the 27-kev X rays pass. When the source is not in the exposure position, the dose rate at the capsule surface is less than 0.1 mr/hr.

The source may be used in making roentgenograms of industrial equipment[2] (Fig. 3) and of teeth (Fig. 4) and bones (Fig. 5).[3-7] The exposure time required with

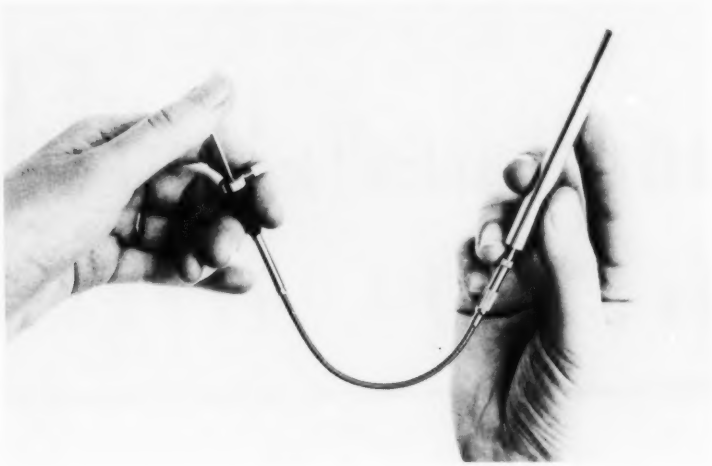

Fig. 1 Iodine-125 X-ray-source apparatus.

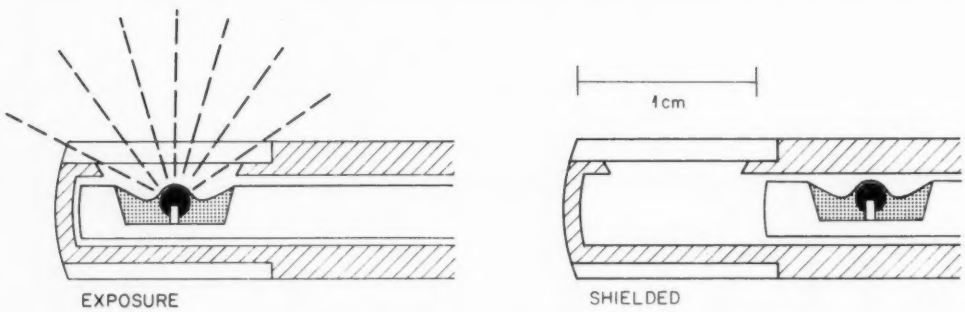

Fig. 2 Radiation source in the exposure and shielded positions.

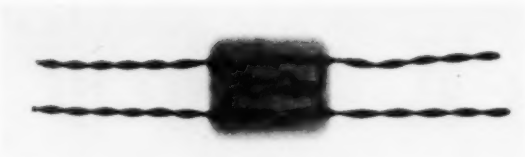

Fig. 3 Diode bridge-type rectifier, X-rayed with a 35-μc ^{125}I source at 100 mm. Polaroid 57 Land film with a special lighting screen.

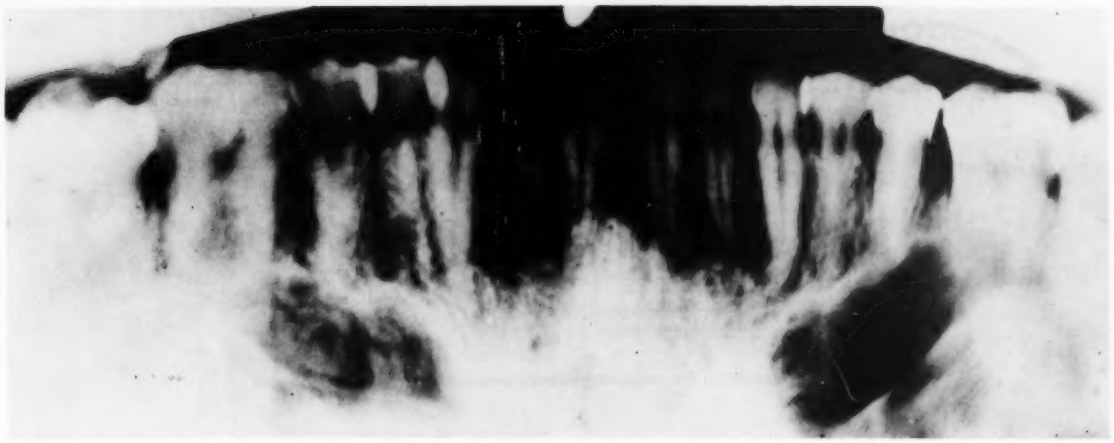

Fig. 4 An X-ray of upper teeth made with a 0.3-curie ^{125}I source, using a bite device; 20 sec exposure time; nonscreen medical film, Cea Test-x.

Fig. 5 An X ray of hand made with a 1.96-curie ^{125}I source at 12 in. Intensifying screen used; 2 min exposure (9.5-mr dose); Kodak Royal Blue X-ray film.

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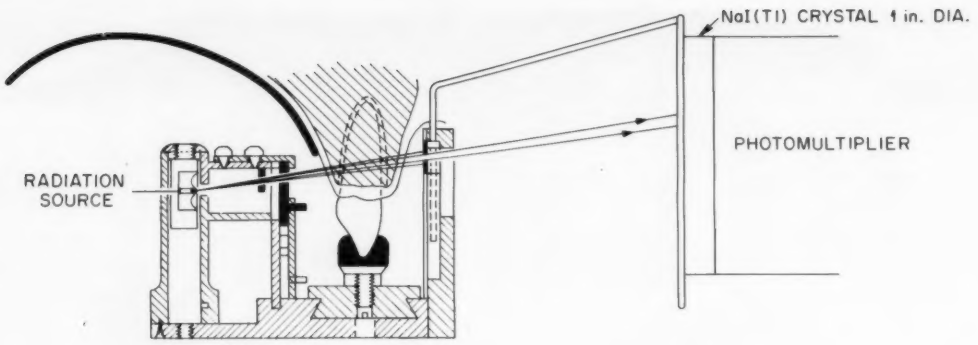

Fig. 6 Arrangement for measuring mineral content of alveolar bone in vivo.

a source of a given strength is dependent on, among other factors, the type of film and the source-to-film distance. For a 0.5-curie source, e.g., the exposure times are

Object	Film	Source-to-film distance, cm	Exposure time, sec
Teeth	Kodak ultraspeed dental	6	10 to 20
Hand	Kodak Kodirex	20	~40
Vacuum tube	Medical X ray	20	~30

Other uses include determination of the mineral content of bone[8] (Fig. 6) by measuring the amount of radiation transmitted and determination of thickness or density by measuring the X radiation backscattered or transmitted, e.g., the thickness of a fiber.[9]

The preparation of ^{125}I and of radiation sources from it has been described in detail elsewhere.[3,4,10]

(MG)

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New Technique for Rapid Preparation of ^{99}Mo for ^{99m}Tc Generators

A solvent extraction process for recovering ^{99}Mo from neutron-irradiated ^{235}U has been developed at Oak Ridge National Laboratory—a process that requires only one-fourth the time required by the previous ion-exchange technique. Commercial radiopharmaceutical producers sorb the ^{99}Mo product on an ion-exchange material, from which the 6-hr daughter product ^{99m}Tc can be eluted for use in nuclear medicine.

(MG)

A Conceptual* Design of a Continuous Thermal-Diffusion Plant for ^{85}Kr Enrichment

By K. H. Lin

The conceptual design of a continuous multicolumn thermal-diffusion plant for enrichment of ^{85}Kr was described in a recent report (ORNL-4372). The design was developed for the purposes of assessing its economic feasibility and identifying important design problems. Feed material containing 4% ^{85}Kr is to be enriched through the cascades of thermal-diffusion columns to a product of 45% ^{85}Kr at a rate of 100 thousand curies/year. About 150 thousand curies of feed will be required per year.

Two alternative systems—each consisting of two cascades—were compared; the systems are identical except for the intercascade flow arrangement. In one, the effluent, rich in heavier isotopes from the first cascade, is fed to the second cascade. In the other system the lighter isotope mixture from the first cascade is fed to the second.

Details of the design may be found in the original report. A single column is shown in Fig. 1. (MG)

*The plant now in use was described by W. R. Rathkamp, The Enrichment of Radioactive ^{85}Kr, *Isotopes and Radiation Technology*, 6(4): 347-352 (Summer 1969).

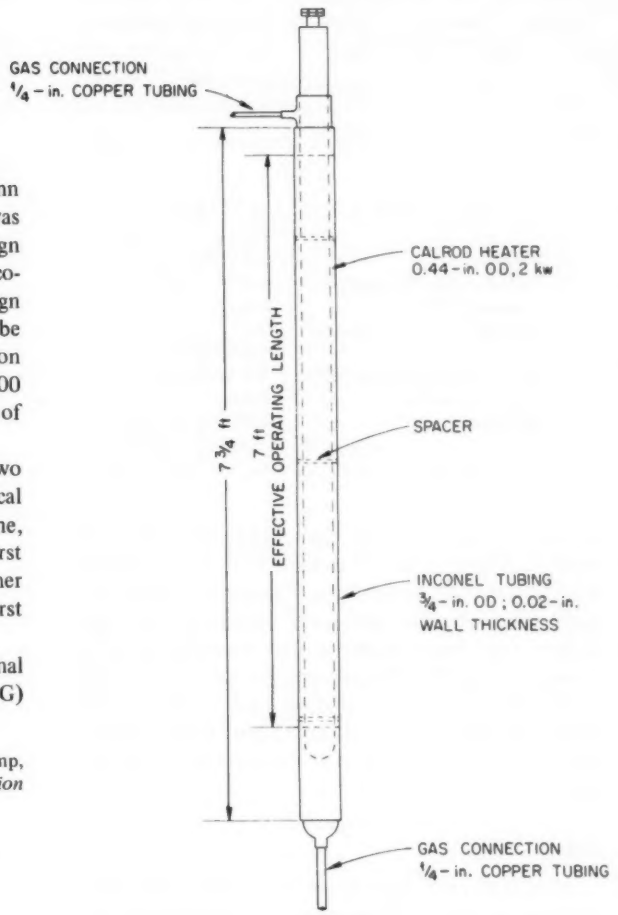

Fig. 1 ORNL thermal-diffusion column. (Not to scale.)

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Thermal Analysis of Promethium Oxalate

By C. E. McNeilly and F. P. Roberts*

As the rarest member of the "rare" earths, promethium does not occur at all in nature and is strictly a by-product of nuclear fission occurring in plutonium production and power reactors. The most abundant isotope, ^{147}Pm, has a half-life of 2.62 years and decays by beta emission to ^{147}Sm.

It has been realized for several years that ^{147}Pm has certain characteristics which make it desirable for use in isotopic heat sources (Table 1).

Table 1 Characteristics of ^{147}Pm

Half-life	2.6 years
Watts per gram of element	0.33
Compound form	Pm_2O_3
Watts per gram of compound	0.27
Density	6.6 g/cm^3
Power density (compound)	1.8 watts/cm^3
Type of radiation	Beta
Shielding	Minor: 1 in. lead for 1-kw source

As with most rare earths, the sesquioxide is the most stable and hence the most suitable form of the element for heat-source applications. However, since the promethium must be chemically separated from other fission and decay products and precipitated as a hydrated oxalate, it is necessary to calcine it to the oxide. Both differential thermal analysis (DTA) and thermogravimetric analysis (TGA) were used in studying this calcination, and it is these results which are presented here.

Experimental

For protection against the biological hazard from the beta-emitting promethium, all analyses were performed in totally enclosed glove boxes. The DTA apparatus was constructed so as to facilitate operation and maintenance in the glove box. The furnace is heated with a Pt–40 Rh winding, uses Pt–10 Rh thermocouples, has a platinum liner in the furnace tube, and employs d-c power to reduce electrical "noise" in the differential signal (this permits a differential signal sensitivity of 3 μv/in., although 10 μv/in. is ordinarily used). The TGA system includes a standard Ainsworth semimicro automatic balance (10-mg full-scale sensitivity) and a platinum furnace.

Atmospheric control is available for both DTA and TGA, although most of the work was performed in air. Sample sizes were standardized at 17 ± 1 mg for both DTA and TGA.

Because of their proximity in the periodic chart, it was anticipated that promethium and samarium oxalates would decompose similarly. However, the DTA patterns are not at all similar (Fig. 1). The promethium oxalate apparently loses all its water of hydration at once (and at a lower temperature) compared to three distinct stages for samarium oxalate. Furthermore, there was no indication whatsoever of promethium oxalate decomposition, which shows up so predominantly with samarium and most other rare-earth oxalates.[1,2]

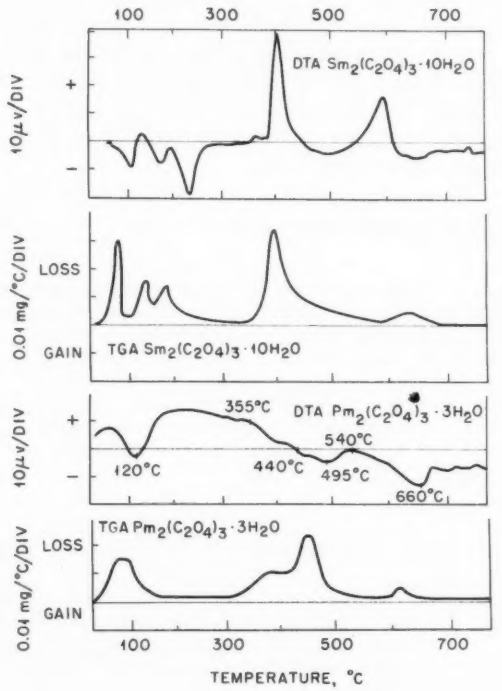

Fig. 1 DTA and TGA of samarium and promethium oxalates.

*Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Wash. 99352.

To assess any self-radiolytic effects, since TGA on 3-week-old samples showed only about a 32 wt.% loss instead of the theoretical 44 wt.% from $Pm_2(C_2O_4)_3 \cdot 3H_2O$, DTA and TGA were performed regularly on newly prepared promethium oxalate over a period of several weeks.

Results

Figure 2 shows DTA and TGA results for material that was dried at about 50°C to constant weight (about 2 hr) in the apparatus. Analysis of the TGA results shows that by calculating back from the Pm_2O_3, which is obtained at about 660°C by endothermic decomposition of $Pm_2O_2CO_3$, the starting material was very nearly $Pm_2(C_2O_4)_3 \cdot 3H_2O$ (which appears to be the normal hydrated form) and that $Pm_2(C_2O_4)_3$ and $Pm_2O_2CO_3$ are intermediate decomposition products. However, the true $Pm_2(C_2O_4)_3 \cdot 3H_2O$ exists for only a few hours after gentle drying of the precipitate.

The DTA for this material also shows the expected oxalate exothermic reaction, although it is at a higher temperature than expected from the results with samarium oxide.

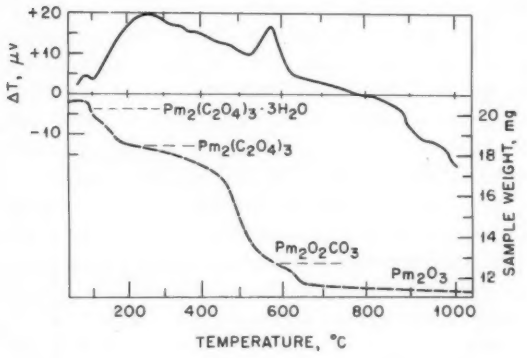

Fig. 2 DTA and TGA of freshly prepared promethium oxalate.

Figure 3 shows the results on the same material 4 days later. This material had been dried at 50°C for about 48 hr. It is difficult to tell how much of the change is due to excessive drying and how much to radiolytic damage. However, since the DTA and TGA runs during the next few days did not show extensive change, it can probably be assumed that the effect was primarily thermal.

The longer-term time effect of the radiolytic damage is shown in the DTA results in Fig. 4. The

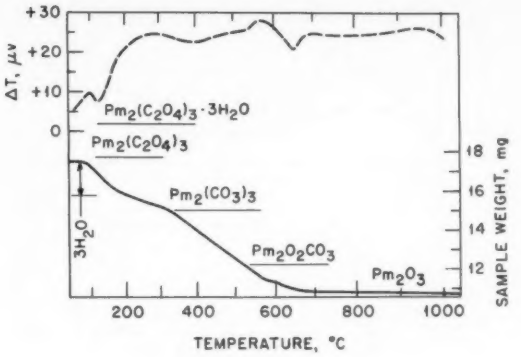

Fig. 3 DTA and TGA of 4-day-old promethium oxalate.

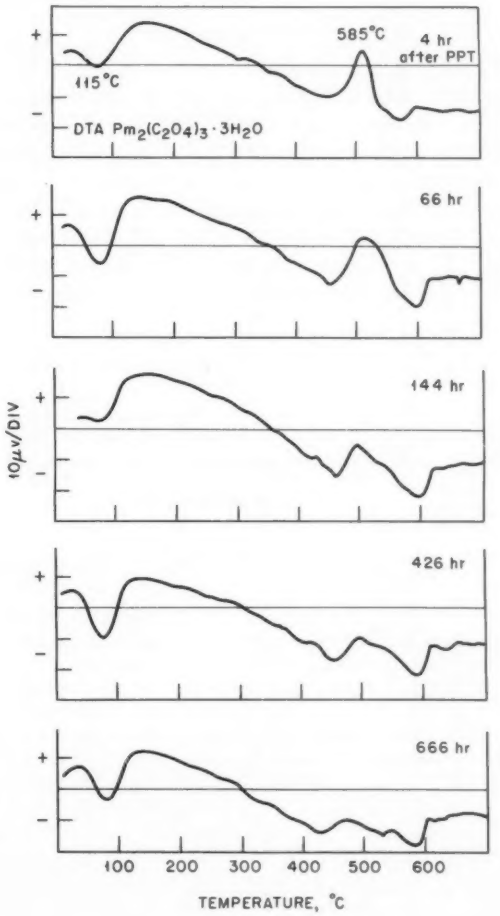

Fig. 4 Effect of time on decomposition of promethium oxalate.

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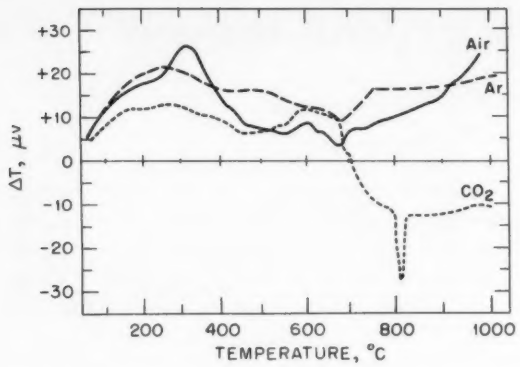

Fig. 5 Effect of atmosphere on DTA of promethium oxalate.

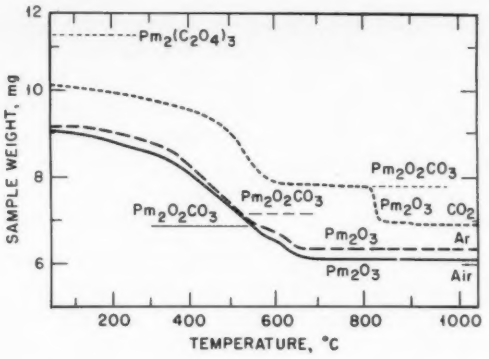

Fig. 6 Effect of atmosphere on TGA of promethium oxalate.

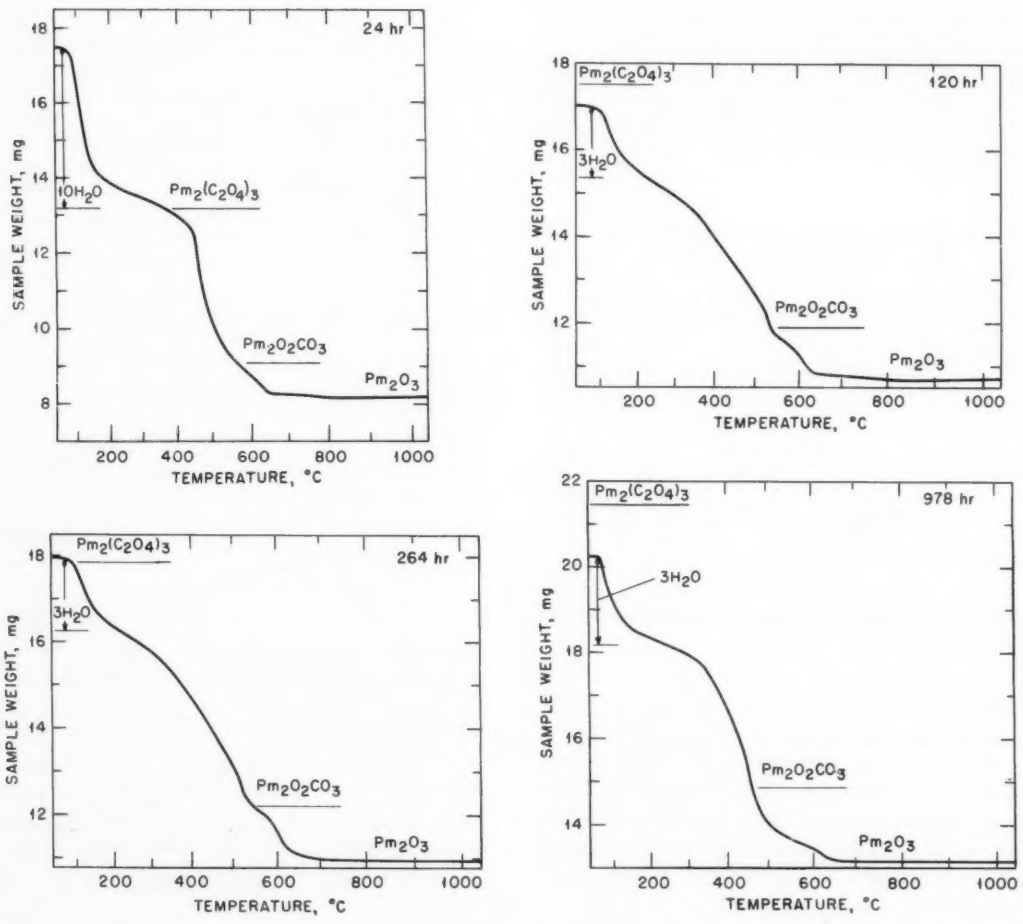

Fig. 7 Effect of time on TGA of promethium oxalate.

endotherm due to water remains approximately the same at about 120°C, while the oxalate exotherm at about 580°C decreases in magnitude. An additional endotherm appears in the vicinity of 655°C, which appears to be due to the decomposition of the $Pm_2O_2CO_3$. This is supported by results shown in Figs. 5 and 6. From Fig. 5 it is readily apparent that a carbonate decomposition which occurs at about 815°C in a CO_2 atmosphere is probably the same as that which occurs at 655°C in air.

The calculations based on the data shown in Fig. 6 indicate that the compound $Pm_2O_2CO_3$ is quite stable in CO_2 up to 815°C, where it decomposes readily to $Pm_2O_3 + CO_2$. (It should be pointed out that this material had been held in a vacuum desiccator for about 18 hr before the DTA and TGA were performed. This treatment is sufficient to remove all water, including the chemically combined water. Because the chemically combined water normally comes off at about 120°C, it would seem to indicate that this water is not very tightly bound in the lattice.)

Figure 7 shows a series of TGA results obtained over a period of 978 hr (approximately 41 days), starting with freshly precipitated promethium oxalate. The first curve is for 1-day-old material that had been only air dried and so contained excess water. The formula for this material calculated to be $Pm_2(C_2O_4)_3 \cdot 10H_2O$. The results obtained at 120 and 264 hr were for material dried at 50°C for 48 hr and show the more usual 3 moles of water. They also illustrate the fact that the sample is no longer pure oxalate and that $Pm_2O_2CO_3$ appears to be an intermediate product, forming at approximately 520°C. (Note that this is slightly lower than the 580°C exotherm of DTA.) The sample analyzed at 978 hr had been only air dried since precipitation and in addition to the 3 moles of water shows the presence of stable $Pm_2O_2CO_3$.

In some cases, calculations indicate that the starting material must have had a lower average molecular weight than the oxalate. For example, for the 978-hr run, the weights of the starting material and final product suggested an average molecular weight of 525 instead of 612, which would indicate a mixture of 62 wt.% $Pm_2(C_2O_4)_3 \cdot 3H_2O$ and 38 wt.% $Pm_2O_2CO_3$.

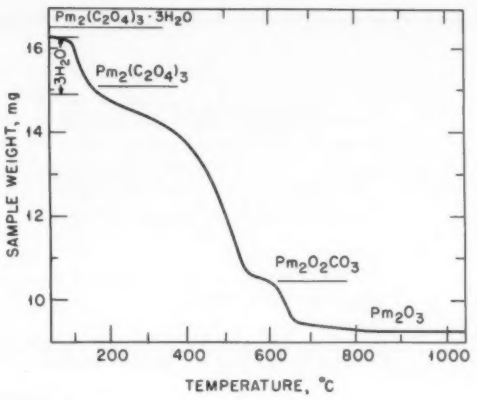

Fig. 8 The TGA of promethium oxalate stored in sealed container for 600 hr.

That such a conversion can take place is evidenced to some extent by the information shown in Fig. 8. This material was kept in a sealed container after precipitation and drying and was run about 600 hr later; however, it appears to be essentially $Pm_2(C_2O_4)_3 \cdot 3H_2O$. On the other hand, the material represented in Fig. 7 had been periodically opened to remove samples. The radiolytic conversion of the oxalate to an oxycarbonate follows the reaction

$$Pm_2(C_2O_4)_3 + \frac{3}{2}O_2 \xrightarrow{\beta} Pm_2O_2CO_3 + 5CO_2 \uparrow$$

As the partial pressure of CO_2 in the container increases, this reversible reaction is retarded. (PSB)

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*Radioisotope Production in the IAN-R1 Reactor**

Standard methods for producing radioisotopes are compiled (64 references), with special interest in the feasibility of producing isotopes in the Colombia 10^{11}-n/(cm^2)(sec) reactor. Reactions considered are the (n,γ) for ^{131}I, ^{24}Na, and ^{198}Au, using ^{130}Te, ^{23}Na, and ^{197}Au, respectively; (n,p) for ^{14}C, ^{35}S, and ^{32}P, using ^{14}N, ^{35}Cl, and ^{32}S, respectively; and uranium fission for production of ^{137}Cs as well as ^{131}I. Details of chemical separation are included. (MG)

*In Spanish, by E. A. Paez and S. J. Nassiff, Instituto de Asuntos Nucleares, Bogota, Colombia, Report IAN-Q-6, 1966.

^{123}I: New Production Method

A new method for producing ^{123}I has been announced by the Bureau of Radiological Health of the U. S. Department of Health, Education, and Welfare. The method was developed by Dr. Vincent J. Sodd of the Nuclear Medicine Laboratory of the Bureau of Radiological Health in Cincinnati and Dr. James W. Blue of the National Aeronautics and Space Administration's Lewis Research Center in Cleveland, Ohio. They obtain ^{123}I by decay of the parent radionuclide ^{123}Xe, which is produced by alpha-particle bombardment of 95.4% enriched ^{121}Te in a continuous-flow cyclotron target. During bombardment, a recirculating flow of helium flushes the xenon out of the target and into a liquid-nitrogen trap, where it is frozen out. The trapped xenon decays with a 2-hr half-life to ^{123}I by positron emission. After the ^{123}Xe has decayed a sufficient length of time, the trap is purged with helium to remove undecayed radioxenons. The ^{123}I is then flushed out with 0.5*N* NaOH. The product contains less than 0.01% ^{124}I. Since the ^{123}Xe may be shipped to the user, there is somewhat less loss of ^{123}I in transit than when the product ^{123}I itself is shipped. An application for a patent on the process has been filed by the National Aeronautics and Space Administration. Iodine-123 is also produced by proton bombardment* of 80% enriched ^{123}Te and by alpha-particle bombardment of natural antimony† and of 98.4% enriched ^{121}Sb.‡ The advantages of this iodine isotope over other radioactive isotopes of iodine for diagnostic purposes were discussed in an earlier state-of-the-art review.§ They include:

1. Low radiation dose to patient due to (*a*) short half-life, (*b*) absence of particulate emissions during decay, and (*c*) low dose-rate constant.
2. High yield of detectable photons due to (*a*) high abundance of both X and gamma rays, (*b*) low tissue absorption of gamma rays, and (*c*) high efficiency of detection of X and gamma rays.
3. Simplified collimation and counting due to (*a*) convenient energies of the X and gamma rays and (*b*) simple or "clean" gamma-ray spectrum.
4. Short half-life which allows for (*a*) repetitive testing and (*b*) low interference with other radioisotope tests.
5. Estimation of target depth by measuring both the X- and gamma-ray counts.
6. Better counting statistics and/or reduced patient exposure.
7. Automatic, rapid cleanup of any spills. (MG)

*H. B. Hupf, J. S. Eldridge, and J. E. Beaver, Production of ^{123}I for Medical Applications, *Int. J. Appl. Radiat. Isotop.*, **19**: 345-351 (1968).

†A. W. G. Goolden, H. I. Glass, and D. J. Silvester, The Choice of a Radioactive Isotope for the Investigation of Thyroid Disorders, *Brit. J. Radiol.*, **41**: 20-25 (1968).

‡V. J. Sodd and J. W. Blue, Production of ^{123}I by the ^{121}Sb(a,2n)^{123}I Method, submitted to *Int. J. Appl. Radiat. Isotop.*

§B. A. Rhodes, H. N. Wagner, Jr., and Martha Gerrard, ^{123}I: Development and Usefulness of a New Radiopharmaceutical, *Isotop. Radiat. Technol.*, **4**(3): 275-280 (1967).

Section
II
Isotopes and Radiation Technology

Isotope Technology Development

Measuring Moisture with Gamma Radiation

By F. E. McKinney

Abstract: *The literature on applications of gamma radiation to the determination of moisture content in soil, peat, wood, ceramic materials, construction materials, meat, and textile fibers is reviewed briefly. Activities used were 1 mc to 10 curies of ^{60}Co and 5 mc to 251 mc of ^{137}Cs.*

Determining the amount of moisture in a given substance is a necessary part of many physical and chemical processes. Neutron sources are commonly used for this purpose, especially in soil studies. There are many references in the literature to neutron moisture gages, and at least one commercial device is available in the United States. *Isotopes and Radiation Technology* has reported on the development[1-3] of these gages. The neutron moisture gage is based on the principle that hydrogen is a good moderator of neutrons, i.e., a neutron moisture gage actually measures the amount of hydrogen in a substance. However, many substances have hydrogen as part of their chemical makeup; neutron measurements would be in error for these materials. Some researchers have therefore used gamma radiation to measure the difference in densities between a dry sample and a test sample of a substance, with the assumption that the difference in densities is the result of the difference in water content. The literature on the applications of gamma-emitting radionuclides to the measurement of moisture content in various substances is briefly reviewed here.

Soil

Unger[4] reviewed 18 different methods of indirect determination of soil moisture using radionuclides, 7 of which were gamma moisture methods (Fig. 1). All seven methods are based on the assumption that changes in soil density with time are actually changes in moisture content. Method 1 in Fig. 1 has the disadvantage of being a permanent installation; many radioactive sources would be required for research over a large area. Method 2 uses a guide tube for insertion of the source below ground and would be more economical than method 1 since only one source would be required. Method 3 uses a device that is driven into the ground at each measuring point rather than lowered into a previously installed tube. In method 4 the

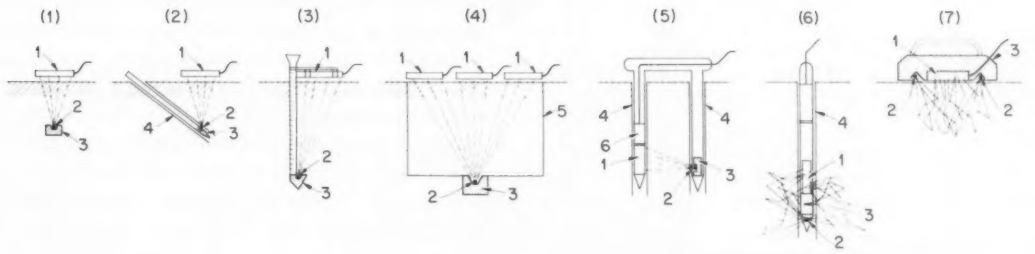

Fig. 1 Summary of gamma moisture-measurement configurations. (From Ref. 4.) Legend: 1, detector; 2, gamma-radiation source; 3, lead shield; 4, guide tube; 5, lysimeter; and 6, amplifier.

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gamma source transmits its radiation through a lysimeter to multiple detectors. Method 5 uses a forked probe with the detector output run through an amplifier; the gamma radiation is measured horizontally through the soil as the device is raised and lowered inside its guide tubes. Methods 6 and 7 differ from the first five in that scattered radiation rather than transmitted radiation is measured. Method 7 uses two gamma sources.

At a recent Czech symposium,[5] two of four papers on soil-moisture measurement described the use of gamma sources[6,7] and reported comparisons with neutron sources.

At a 1960 conference, Danilin[8] —credited by Unger with pioneering in gamma moisture measurement—reported on two measurement arrangements. One is similar to method 2 in Unger's summary. The addition of a second tube below the first allows a set of three measurements to be taken at a single point with one gamma source (Fig. 2). Danilin's second method is a simplified version of method 5. He also reported economic data. Comparison with the usual method of measuring soil moisture (sampling, drying, and weighing) showed that the gamma method used 1 less worker to make the measurements (15 fewer workers to obtain data from five fields in the same length of time), gave immediate results (the usual method takes 10 to 15 hr), and required less energy to make the measurement because electricity is not required for the operation of desiccation cabinets. Danilin estimated savings of 400,000 rubles/year. Over a set of 160 measurements from three different agricultural stations, the maximum difference between the sampling, drying, and weighing technique and the gamma technique was only 4% in two cases, and 88 to 95% of the measurements differed by less than 2%.

At the same conference, Zavel'skii[9] reported observations of soil-moisture dynamics in the aeration zone using gamma absorption. He used a system similar to Danilin's two-borehole method, with a counter in one tube and the gamma source in the other. Zavel'skii used an uncollimated gamma source with appropriate correction for scattering. He reported an accuracy to within 3%.

At least six researchers have used gamma absorption to measure water content of soil in the laboratory. Massaux and Bentz[10] used the device in Fig. 3 to achieve results accurate to within 0.5% of theoretical.

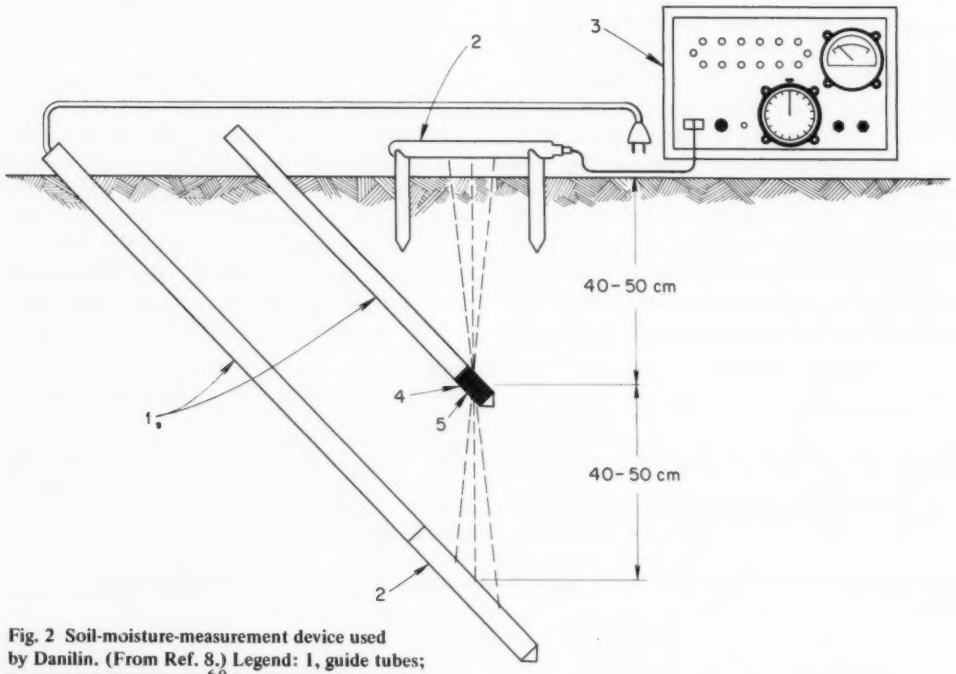

Fig. 2 Soil-moisture-measurement device used by Danilin. (From Ref. 8.) Legend: 1, guide tubes; 2, counters; 3, scaler; 4, ^{60}Co gamma source; and 5, lead shield.

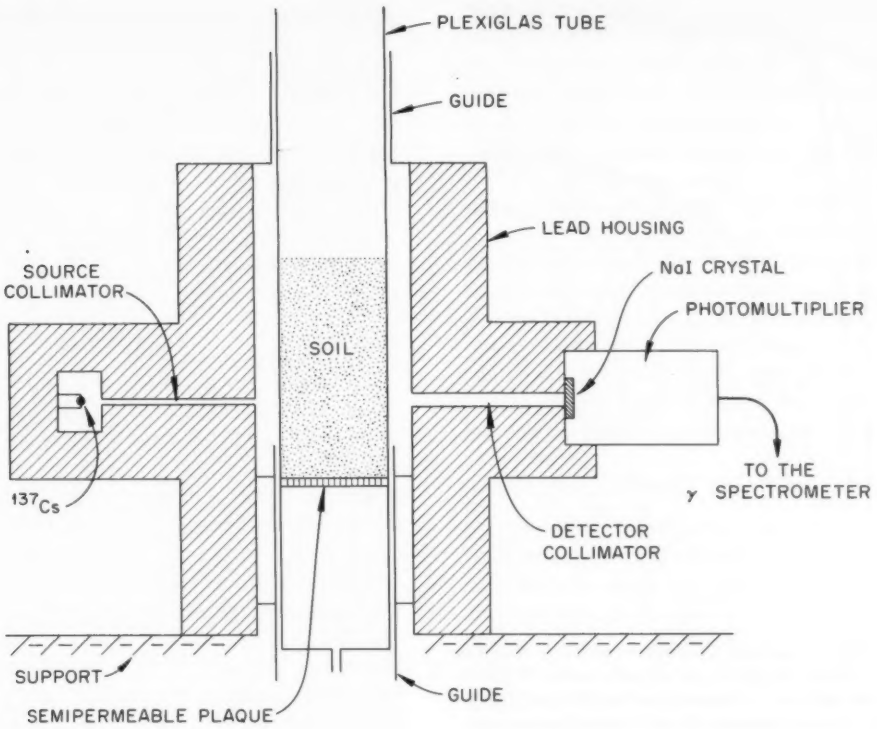

Fig. 3 Measurement of water content in soil sample by gamma transmission method. (From Ref. 10.)

Gurr[11] pointed out that the soil density in the column must be constant, i.e., the soil must not swell or shrink as the moisture content changes; otherwise, errors will result. His results are given in Table 1. The greater error in measurement toward the bottom of the column he ascribes to increasing density of the sand in the lower part of the column.

Ferguson and Gardner[12] claim that all other methods of determining soil water content—including the neutron-scattering method—suffer from one or more of the following limitations: lack of accuracy, poor resolution, insufficient range, lag in following water-content changes, disturbance of the system through addition or removal of water, or destruction of the system. The major limitation of the gamma method is that changes in bulk density affect its accuracy. Since water is a poor absorber of high-energy gamma rays, these workers advocate the use of ^{137}Cs (0.662 Mev) rather than ^{60}Co (1.17 and 1.33 Mev). Their results are shown in Fig. 4.

Kirkham, Rolston, and Fritton[13] used 251 mc of ^{137}Cs to measure changes in moisture of a two-

Table 1 Comparison of Gamma-Ray Measurements and Sampling of Water Contents in a Column of Sand

Depth,* cm	Density, g/cm^3	Corrected counts (in 300 sec)	Water content		
			Calculated from counts, g/g	Sampled, g/g	Error
1.25	1.641	519,047	0.073	0.081	−0.008
2.5	1.630	524,849	0.074	0.074	0
3.75	1.641	510,980	0.080	0.080	0
5.0	1.630	517,511	0.081	0.079	+0.002
6.25	1.633	514,811	0.082	0.080	+0.002
7.5	1.632	513,517	0.084	0.082	+0.002
8.75	1.615	520,118	0.088	0.083	+0.005
10.0	1.642	493,683	0.097	0.088	+0.009
11.25	1.617	512,385	0.094	0.085	+0.009
12.5	1.640	488,103	0.104	0.103	+0.001

*From top of column.

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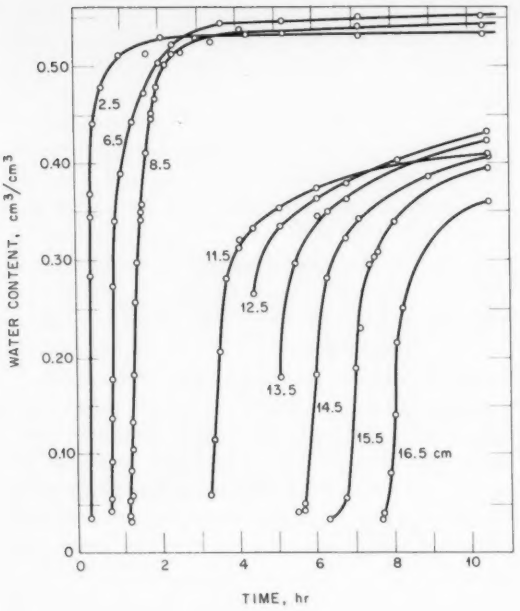

Fig. 4 Water-content variation with time at different distances from a water source, measured with a gamma source in a layer of Salkum silty clay loam. The break between 8.5 and 11.5 cm is caused by a layer of aggregates 9 cm from the water source. (From Ref. 12.)

dimensional soil model and thus evaluate the benefits of mulching to prevent evaporation. Reginato and van Bavel[14] measured moisture in 26 soil samples, using the device shown in Fig. 5. A total of 20,000 counts was recorded at 10 locations in each sample. These researchers claimed an accuracy of 0.010 g/cm^3 for water contents from 0.050 to 0.400 g/cm^3 at a dry density of 1.300 g/cm^3. The device differs from other gamma moisture-measurement arrangements by having a magnesium bar built in to use as a calibration standard. Reginato and van Bavel point out that, although the method may indicate moisture changes precisely, the absolute moisture content may be subject to error. These researchers state that the principal justification for using gamma attenuation to measure moisture is its superior resolving power.

As part of a study using radioisotopes as tracers of moisture in soil columns 90 cm long, Shalhevet and Yaron[15] used a 19-mc ^{137}Cs gamma source to measure moisture content in columns being dried to simulate the irrigation–evaporation–transpiration cycle. Table 2 compares the gamma measurements with gravimetric measurements.

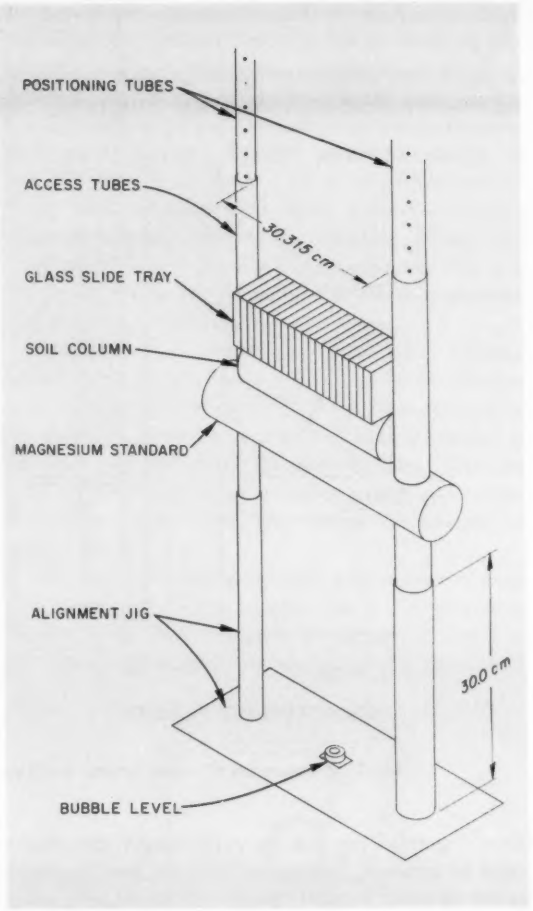

Fig. 5 Laboratory setup for calibration of gamma transmission apparatus. Gamma emitter is in tube on right and detection system in tube on left. (From Ref. 14.)

Table 2 Estimation of Moisture Content Calculated from Gamma Attenuation and Measured Gravimetrically (Adapted from Ref. 15.)

Time of drying, min	Volumetric moisture content		
	Calculated, %	Measured, %	Difference, %
0	24.9	25.1	−0.2
22	21.0	21.8	−0.8
45	18.4	18.1	0.3
68	15.5	15.1	0.4
Air dry	2.85	2.00	0.2

Other Substances

Peat

Volarovich, Minkov, and Churaev[16] described two field devices based on gamma radioscopy for measuring moisture in peat deposits and reported observations at Gusevskoe and Yuzhnoe Alferovskoe in 1957, at Radovitskii Mokh in 1958, and Orshinskoe in 1958–1960. The gamma moisture gages greatly reduced difficulties of observation and increased accuracy. Measurement error in relative peat moisture did not exceed ±0.2 to 0.3%.

Ceramic Materials

Cermak[17] compared moisture determination in raw ceramic materials by neutron and by gamma-ray absorption. He reported an accuracy of ±1% for neutrons and of ±6% for gamma radiation, with a treatment of the factors such as grain size and density that caused errors.

Holzhey and Schwarzlose[18] measured moisture content in extruded ceramic material, using a gamma device calibrated by three different sampling methods. Calibration samples were taken from the very middle of a cross section, at three different positions in a line with the radiation path, and in one continuous slice along the radiation path. The last method gave the best calibration of the gamma device.

Cement and Construction Materials

Saito and coworkers[19] measured water content of raw cement slurry. For optimum operation, water content should be about 40%—high enough to make flow easy but low enough to allow proper operation of the rotary kiln, i.e., without having to use excess heat or lowering the quality of clinker. They reported that their device could measure a 0.5% change in water content.

Change in moisture content of construction materials, especially foamy silicate, was measured with a gamma source by Polozova and Reizman.[20] Foamy silicate is a form of foamy cement developed in the Soviet Union in which the cement is replaced by a mixture of lime and sand. Differences between values obtained by the gamma method and by the usual method of drying and weighing averaged about 1.5%. The device was used for about a year.

Wood

Sakamoto and Iizuka[21] attempted to detect rotten wood in standing trees and wood materials by a gamma transmission method, assuming that rotten wood would contain less moisture. In a grove of 34 trees, they claimed a 93% accuracy in locating rotten portions of the trees. The apparatus was also used to measure relative water contents on some green wood disks, which were also measured by the absolute drying method. Distribution curves of absorption counting rates and of water contents corresponded well.

Jokel[22] determined the absorption of gamma rays by lime wood and showed that absorption depended on the moisture content of the wood. However, he determined that the regression coefficients of dry and moist wood having the same density are not significantly different, and Jokel concluded that in determining the density of moist wood with a gamma method the moisture content can be neglected.

Textile Fiber and Meat

Recent work by Hatcher, Lyons, and Sunderland[23] has been directed toward determining moisture content in a material under transient drying conditions. Since the sample and physical test conditions remain constant, change in radiation attenuation is directly comparable to moisture removal from the sample. The gamma method is superior to the neutron method in this case for two reasons: attenuation of neutrons in an organic product is a function of the hydrogen in the product rather than of the moisture present, and neutrons are more likely than gamma rays to activate the sample. These researchers point out that a scintillation crystal is superior to other photon detectors in measuring moisture because the crystal response is a more sensitive measure of the energy of the photons reaching it. Changes in gamma attenuation and, thus, changes in moisture content are more quickly detected. A pulse-height discriminator should also be used.

Samples measured by Hatcher, Lyons, and Sunderland were arranged so that the moisture flow would be one-dimensional. The meat samples were sealed with plastic wrap and insulated on all sides but one. Either the ends or the circumference of the cylinder of textile fibers was sealed with an epoxy resin. This was done to ensure that the moisture content in the sample did not vary along the path of the gamma rays. Results are shown in Figs. 6 and 7.

Isotopes Used as Gamma Sources

In all cases where the source radioisotope was specified, either ^{60}Co or ^{137}Cs was used. In some cases, where one was used, the other was mentioned as

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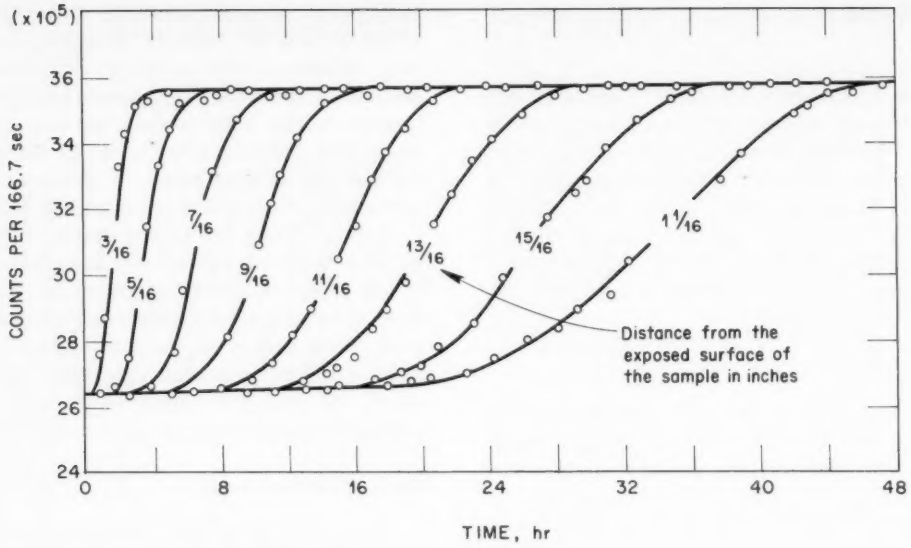

Fig. 6 Variation of moisture in a frozen-meat sample being vacuum dried, measured with a gamma transmission method. (From Ref. 23.)

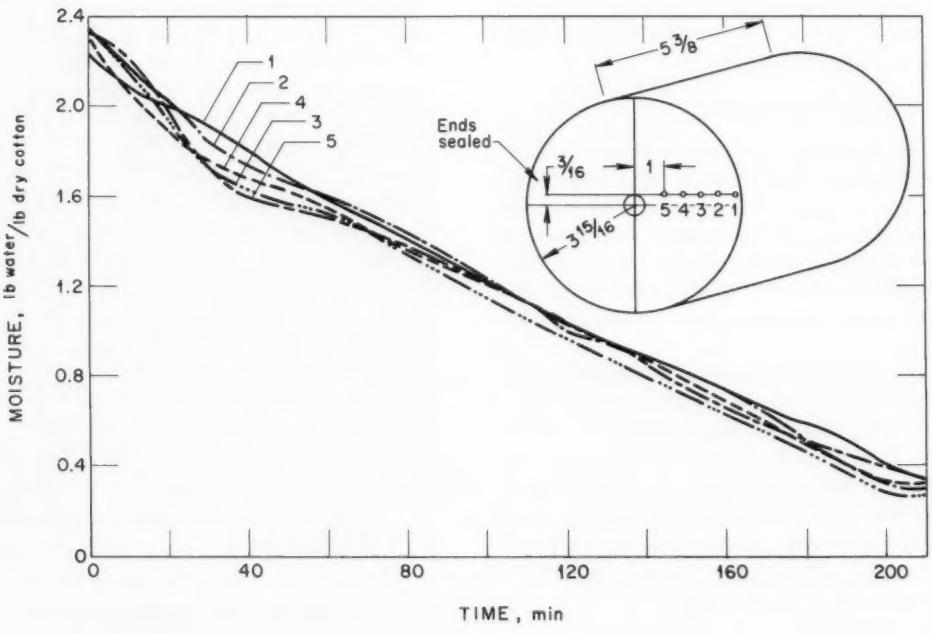

Fig. 7 Variation of moisture in a sample of textile fibers during drying, measured with a gamma transmission method. All measurements in inches; distance between measuring points through cylinder is 0.5 in. (From Ref. 23.)

being a satisfactory alternate. In one paper[12] the lower energy of ^{137}Cs was cited as the reason for its use. Six papers reported using ^{137}Cs in strengths ranging from 5 mc (Ref. 14) to 251 mc (Ref. 15). Eight papers reported the use of ^{60}Co in strengths ranging from 1 mc (Ref. 21) to 10 curies (Ref. 23).

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Nucleonics in Aerospace

Summarized by F. E. McKinney

Abstract: *The 51 papers in the published proceedings of the Second International Symposium on Nucleonics in Aerospace are summarized. The meeting—held in Columbus, Ohio, on July 12–14, 1967—covered gaging, health and safety, detectors, heat sources, nondestructive testing, sources, analytical techniques, and ranging and guidance.*

The Second International Symposium on Nucleonics in Aerospace was held in Columbus Ohio, on July 12–14, 1967. A total of 51 papers, out of 58 presented, are included in the published proceedings.[1] The objectives of the second meeting remained those of the first, namely: exchange information on radioisotope applications in aerospace, foster the use of radioisotopes in aerospace, acquaint military and industry with the potential of radioisotope technology, and report on the state of the art in applications and components. *Isotopes and Radiation Technology* reported on the first symposium in an earlier issue[2] and now is covering the second meeting in detail. There were introductory remarks by E. E. Fowler, Director, Division of Isotopes Development, USAEC, and by Col. D. D. Davis, Director, Air Force Flight Dynamics Laboratory. Two plenary papers followed. The remaining papers are grouped under the eight subject categories of gaging, health and safety, detectors, heat sources, nondestructive testing, sources, analytical techniques, and ranging and guidance.

Plenary Papers

Mayer[3] presented the requirements for semiconductor devices used for gamma detection at elevated temperatures. Germanium *p-i-n* detectors have the disadvantages that only 1 ingot in 10 will have the necessary crystal perfection and lithium-drift mobility and that the detector must be cooled during operation and storage. The overall gamma-detection efficiency of silicon lithium-drift detectors is somewhat low; these units are not satisfactory for spectroscopy applications because the pulse-height distributions are difficult to interpret.

An obvious approach to high-efficiency detection at temperatures approaching 300°C is to fabricate detectors from a wide-bandgap-compound semiconductor where one of the constituents has an atomic number greater than 50. The material should be able to be processed into single-crystal regions of 1-cm^2 area and 5-mm depth, and the number of trapping centers should be less than 10^{12} per cm^3. The applications of such materials will depend on the trade-off between degraded detector performance and the heat shielding and cooling necessary for silicon detectors.

Gamma rays are absorbed in detector material by the photoelectric and Compton absorption processes. A material with as large an absorption cross section as possible should be chosen; therefore, for optimum detector efficiency with minimum crystal growing difficulties, the material should have an atomic number greater than 40, preferably 50. The thickness of a detector is determined by the range of the secondary electrons produced by gamma rays. A 1-Mev electron has a range of about 1 mm in a semiconductor such as germanium which has a density near 5 g/cm^3; therefore, since photoelectrons of this energy can be produced throughout the detector material, the thickness of the detector should be a minimum of 2 mm. The output signal is produced by motion of holes and electrons within the region (called the depletion region) in which the high electric field exists. The width of the depletion region is dependent on the net concentration of donors and acceptors of ions. The maximum net concentration of donors and acceptors can be derived from the applied bias voltage and is approximately equal to 4×10^{11} per cm^3. For high detection efficiency the contacts of the semiconductor material should be noninjecting, i.e., they should not contribute holes or electrons to the semiconductor material, either of which might affect its response to the impinging gamma rays. This is a formidable technical problem with such materials as Si, GaAs, and Ge, and either surface barriers or reverse biased junctions are usually used to minimize injection. The rise time of the pulse is determined by the transit time of the carriers across the depletion region, if trapping is absent. In a useful device the longest practical transit time is about 10^{-6} sec. The minimum carrier mobility for a 2-mm-thick detector is about 100 cm^2/volt-sec. The maximum operating temperature for a specified resolution is determined by the leakage current.

A trapping process is one in which an electron is first captured by a defect center and then has a greater probability of being thermally reemitted than of recombining with a hole. The number and cross section of trapping centers are critical because these centers

can produce a loss of collected charge, degradation in resolution, and a slow component in the transient response. The rate of thermal excitation of trapped carriers (i.e., detrapping) is depth dependent. For shallow centers, where detrapping is fast compared to carrier transit time, the effect is a reduction in mobility of carrier. For deep centers a slow component is introduced in the transient response. The detrapping time is exponentially dependent on temperature.

Mayer next considers the special case where carriers are formed close to one electrode and presents the analytical expressions to describe the effect of trapping and detrapping, concluding that the transient response is critically dependent on the values of mean carrier drift time and of mean detrapping time and cannot be represented in a closed form.

In gamma detectors, output pulse height is the result of both hole and electron motion. The contribution of each carrier depends on its travel in relation to the length of the depleted region. For gamma spectrometers the effect of carrier collection through different distances is the primary factor causing a spread of standard deviation in the pulse height. Changes in the ratio of *n*-to-*p* path lengths influence changes in the resolution; thus trapping and detrapping critically affect resolution because of the temperature dependency. In gamma detectors, preventing pulse-height distribution shift with temperature is more important than achieving good resolution. Satisfactory crystals must have *n* path lengths and *p* path lengths greater than 10 times the length of the depleted region, i.e., with less than 10^{13} trapping centers per cm^3. So far this low concentration has not been achieved with semiconductors.

Two approaches within the limits of current technology are to grow crystals in which either the electron path length is more than 100 times the positron path length or the *n* path length is shorter than the length of the depleted region and the *p* path length is very much shorter than the *n* path length.

Mayer concluded that many materials today can meet the conditions of $Z > 40$ and minimum carrier mobility of 100 cm^2/volt-sec, but no material has a maximum net concentration of donors and acceptors and a minimum of trapping centers both with a value of about 10^{12} per cm^3. Several materials have both values at about 10^{16} per cm^3.

Burch[4] reviewed the transuranium production program. Since Seaborg and coworkers discovered plutonium in the 1940's, about 115 isotopes of transuranium elements have been identified. The greatest interest in these isotopes still centers about their physical, nuclear, and chemical properties; however, a market is being developed for some of these isotopes, based on their unusual properties. Californium-252 is being developed as a small intense neutron source; it fissions spontaneously, and 1 g produces $>2 \times 10^{12}$ n/sec. Brookhaven National Laboratory is investigating its use in cancer therapy. Several transuranium isotopes are possibly suitable for use as sources of thermal power: ^{238}Pu, ^{242}Cm, and ^{244}Cm.

Figure 1 shows the overall program for production of ^{252}Cf. The program started with the long-term irradiation of 10 kg of ^{239}Pu at Savannah River Laboratory. Less than 0.3% of this material is finally converted to californium. The intermediate products ^{242}Pu, ^{243}Am, and ^{244}Cm are being irradiated in the High Flux Isotope Reactor at Oak Ridge National Laboratory. The long-range goal of the program—production of gram quantities of californium—may be reached in 1969 or 1970.

Gaging

Hoalst and Webster[5] reported test results of a semiconductor detector nucleonic fuel gage. Fuel gages providing continuous, accurate, and reliable measurement of fuel mass are badly needed inasmuch as malfunctions in fuel gages are *the* leading cause of delay in commercial aircraft and *a* cause for considerable downtime in military aircraft. Two configurations were tested: one of five source–detector pairs and one of ten pairs. The gage system consisted of a radiation source, a lithium-drifted silicon solid-state detector, and associated electronics with output into a summing amplifier. The test tank was an exact replica of a C-133 aircraft wing tank mounted in a test structure capable of simulating combinations of pitch and roll up to ±20°. Radiation sources tested were ^{85}Kr and ^{60}Co. An overall error of ±0.8% of the empty tank and ±1.7% of the full tank was obtained, a result equal to presently available aircraft fuel gages. Most of the system error was judged to be the result of temperature changes, and a thermally compensated system is expected to give better results.

A system for indicating the quantity of engine oil, developed for the giant C-5A cargo aircraft, was described by Callaway.[6] Since the engines of the C-5A are difficult of access both on the ground and in the air, some form of remote measurement was necessary. The complete system consists of one 4-mc ^{85}Kr source, one G–M tube detector, and associated electronics. Another radioisotope, ^{241}Am, was considered because its counting system gave a lower statistical

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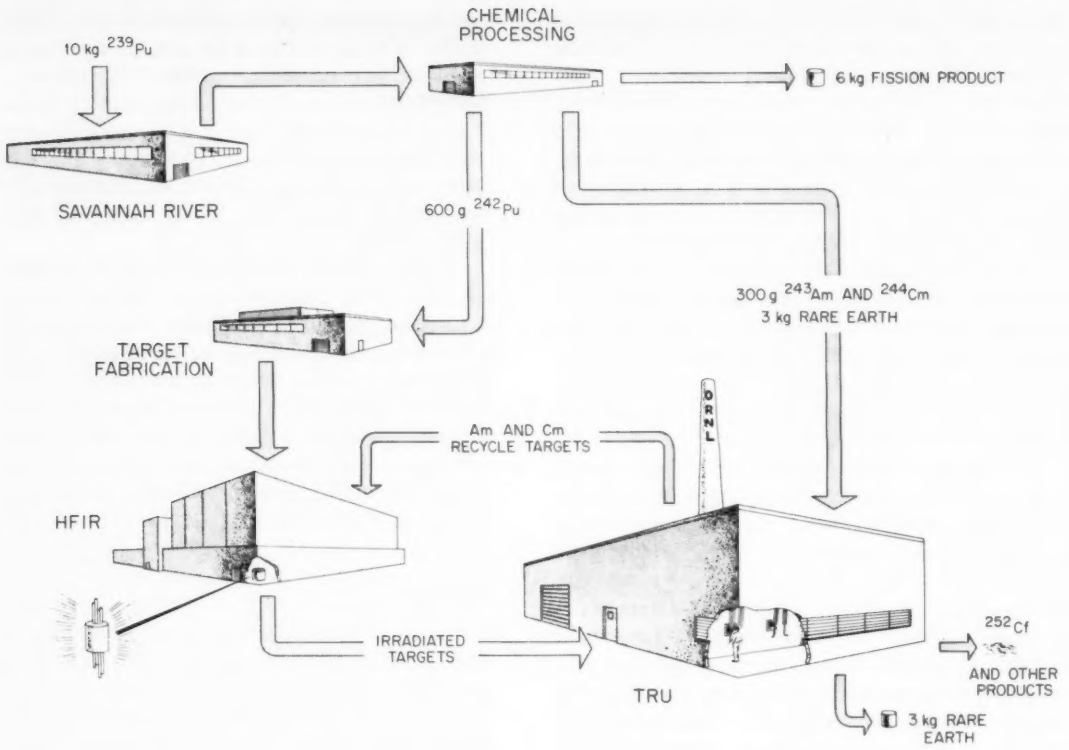

Fig. 1 Heavy-element production scheme.

error and needed less frequent calibration. These advantages were outweighed, however, by the increased hazard during a possible crash and fire. The ^{85}Kr system operates with an accuracy of 3% in normal flight attitudes and 6% during ±20° pitch and ±2° roll. Operating temperatures range from −55 to 232°C. The system life is the half-life of the ^{85}Kr—about 10 years; recalibration is required every 6 months.

Pfeifer, Cho, and Evans[7] examined nuclear techniques for measuring propellant mass aboard orbiting space vehicles. Two regimes of fluid behavior exist: under conditions of acceleration the fluid settles at the "bottom" of the tank, and, under zero gravity conditions, capillary forces and surface tension cause the formation of a single large bubble or a cluster of bubbles. Gaging systems are designed to take advantage of this behavior. Radiation techniques are either mass sensitive or volume sensitive. Volume-sensitive measurements must be supplemented with temperature or pressure measurements to indicate mass. Also, in tanks of liquid oxygen or hydrogen, the difference in size between the radiation source and the tank requires the use of effective sampling techniques.

Mass-sensitive systems may be further classified according to the type of radiation that reaches the detector. Diffusion systems are those systems that detect and analyze all the radiation emitted, whether scattered or direct. Scatter systems detect only scattered photons. Transmission systems detect only primary photons. Volume-sensitive systems may be classified as shadow systems, logic systems, or tracer systems. In the shadow system, matter between an extended line source and an extended detector casts a "shadow" whose geometrical extent allows calculation of the volume occupied by the fluid. Logic systems use numerous source–detector pairs to define the outline of the fluid mass. Tracer systems infer the volume of propellant from the amount of a previously injected tracer present in either liquid or vapor phase.

The three factors that affect the accuracy of mass-measurement systems are buildup, cross talk, and sampling efficiency. Buildup describes the effect of

counting scattered photons with primary photons; if buildup is included to increase the count rate, the correspondence between count rate and absorber thickness is not unique. Cross talk is the complex signal problem in which one detector receives radiation from a number of sources. The sampling scheme must be valid to extrapolate to full data on mass measurement from limited information. Buildup and cross talk affect the diffusion and scattering mass-measurement systems drastically; neither system is generally capable of meeting a 2% accuracy requirement. Transmission systems are better but depend heavily on good sampling efficiency. Cross talk seriously affects volume-sensitive systems, particularly the shadow and logic types. Volume-sensitive systems are generally disadvantageous from an engineering standpoint; shadow and logic systems require extensive internal structures, and the tracer system gives poor results for vented and cryogenic applications. The best overall system is the high-energy, monoenergetic, gamma-transmission system.

The fundamental source of error is related to deficiencies in the sampling technique. The tank is sampled along its length, parallel to its axis. The mass in the tank is approximated by a summation of the cylinders measured, i.e., by the number of beams passing through the tank. The beam is attenuated by the matter in its path. Because there are only a certain number of beams, each measured path represents the matter around it; this is satisfactory since propellant distribution and orientation is not random. However, the sampling beams must be arranged in a pattern such that the sampled cylinders are as compact and as uniform as possible. Beam locations were optimized by computer simulation of the spherical-bubble environment; an array of 12 locations appeared sufficient to provide 2% system accuracy.

Experimental tests were conducted using a 20-mc ^{137}Cs gamma source. The average error was 1 to 2%, and the maximum was less than 3% for 16 tests on foam spheres submerged in tanks.

Three factors limit the range of gamma transmission through hydrogen: flux at the detector through an empty tank must not exceed 5 mr/hr, tank length may exceed dynamic range, and background when the cylinder is full may lower statistical accuracy. The range for ^{137}Cs is about 700 cm, for ^{60}Co about 900 cm. Locating the source at the middle of the tank and the detectors at both ends would permit a maximum length of about 1800 cm to be measured.

Nucleonic measurements of cryogenic density and quality of hydrogen were reviewed by Scheiwe and Cho.[8] Specifically, two gages—one used to measure density of liquid hydrogen (LH_2) at NASA's Marshall Space Flight Center and the other used to measure the hydrogen vented during flight from the Saturn S-IVB stage—were described, and the results from their use were reported. The density gage uses 4 curies of ^{137}Cs as a source, with the radiation collimated into a vertical fan. The apparent accuracy of the device (Fig. 2) was ±0.3% of the density of normal hydrogen; instantaneous accuracy was ±3%. This densitometer was then used by the National Bureau of Standards (NBS) to measure the density of hydrogen slush in a tank that has a diameter nearly 4 times as great as the NASA facility. Accuracy was ±0.5%.

The cryogenic quality gage uses a 1-curie ^{241}Am gamma source. Hydrogen contained in the tanks of the S-IVB stage has to be vented in flight to remove heat energy from the vehicle. Venting superheated vapor is the most efficient procedure. The cryogenic quality was used to measure the relative mass of hydrogen being vented to see if LH_2 as well as vapor was being given off. In-flight TV pictures indicated that the majority of hydrogen being emitted was pure vapor; calculations based on temperature and pressure showed that the vapor was in the superheat region. Since the readout from the nucleonic gage was valid only in the region in which the vapor is saturated with liquid, little information was recovered from its use. Further development of existing instrumentation to provide off-the-shelf units and the design and testing of a flow-rate meter capable of measuring two-phase gas–liquid and liquid–solid cryogenics are needed.

Nichols[9] reported development of a two-phase hydrogen quality meter. The meter is used (1) to obtain the necessary information for calculating the quality, (2) to compute the quality, and (3) to provide a continuous output compatible with telemetry. A ^{204}Tl beta source of 30 mc was used; the source capsule has a variable-width window to allow adjustment for source decay. In operation the instrument measures the density of the vapor and the density of the vapor–liquid mixture and computes the ratio of these values. The density of the vapor–liquid mixture is measured with the beta attenuation method. The LH_2 quality meter met the NASA requirements of accuracy of measurement (i.e., within ±5%). An improved meter is being designed and built in which the pressure transducer is replaced by an alpha-particle attenuation gage.

The use of an isotope to inspect small-arms ammunition to check the level of powder (propellant) was reported by Pollock and coworkers.[10] Four radioactive sources were evaluated: ^{90}Sr, ^{137}Cs,

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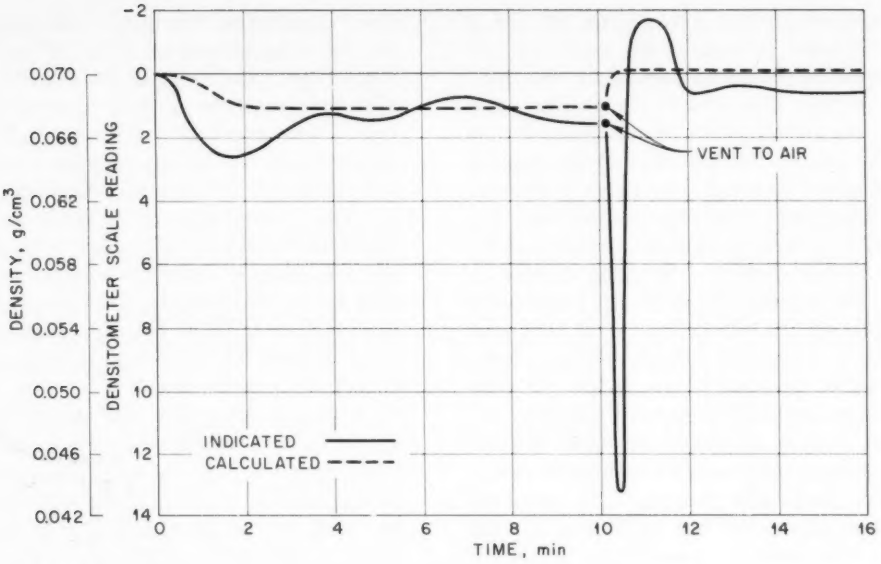

Fig. 2 Accuracy of LH_2 density gage.

^{241}Am, and ^{85}Kr. The strontium and krypton isotopes are primarily beta emitters, but they can also be used to provide bremsstrahlung—which was done in this case. Insufficient propellant causes bullets to fall short, misfire, jam, or even destroy the weapon. Quality control is especially important in today's rapid-fire weapons. The accuracy requirement for this application was ±0.10 in. for three different cartridges—5.56 mm, 30 caliber, and 7.62 mm. Neither ^{85}Kr nor ^{241}Am gave satisfactory results because the collimated radiation beam was not sufficiently intense. Both ^{90}Sr and ^{137}Cs gave good results, with the cesium being the better. Dynamic tests were run on 7.62-mm cartridges at a belt speed of 280 rounds/min. Static tests only were made on the 5.56-mm and 30-caliber cartridges. Pollock and coworkers also developed a mechanical means of measuring and sorting the rounds.

The use of the Mössbauer effect to calibrate measurements of small-amplitude vibrations was described by Netusil and Witherspoon.[11] When testing scale models of space vehicles, a more sensitive method was needed because of the general rule of similitude that vibrational displacement amplitudes in the Angstrom range at frequencies in the kilohertz range are important. The Mössbauer effect observed with ^{57}Fe is best suited for vibration measurements. Cobalt-57, which decays by electron capture to an excited state of ^{57}Fe, was used as the source in these experiments. The study was divided into three cases, based on particular resonance conditions.

A source–absorber combination can be in partial resonance at rest if the chemical environment of the Mössbauer nucleus in each is slightly different. This is the condition of a ^{57}Co/Pd source and a stainless-steel absorber. The source is attached to anything with a variable amplitude of vibration, such as a loudspeaker, and the radiation is transmitted through the stationary absorber to a scintillation counter. Vibrational velocity variations are then detectable in terms of instantaneous count rate. The lower frequency limit was arbitrarily set at the lower limit of the audio spectrum; the upper frequency limit is a function of gating and with improved apparatus will exceed 100 kilohertz. Velocity resolution of 0.01 mm/sec is taken as the lower limit, based on the characteristics of available absorbers. The upper velocity limit is based on symmetry and linearity considerations; here it is 0.1 mm/sec.

Another source–absorber combination can be out of resonance at zero velocity. This is the condition when the absorber has a strong internal magnetic field, which splits and shifts the nuclear energy levels of the Mössbauer nucleus. Multiple calibration points are used in this technique. It is especially useful for small-amplitude electromagnetic, ferroelectric, and piezoelectric transducers. The same frequency limits are

applicable here as they were in the partial resonance case. Velocity limits are 8.4 and 0.25 mm/sec.

In the resonance case the source and absorber exhibit maximum resonance at rest, and vibration decreases resonance. The greater the disturbance, the greater the intensity of the gamma rays transmitted through the absorber and the greater the count rate. The resonance absorption curve is an s-shaped curve that has a useful range only within certain velocity limits since the slope approaches zero for very high and for very low velocities. The upper limit is 1.40 mm/sec, and the lower limit is 0.05 mm/sec. The method has virtually no frequency limits as long as the velocity is within these limits and the wave form is perfectly sinusoidal.

From the studies of Netusil and Witherspoon,[11] a prototype calibration system is being built. It will operate at frequencies above 1 MHz and velocities from 0.01 to 8.4 mm/sec, with corresponding amplitudes as small as 1 A. The system will supplement the optical interferometer now used in most laboratories.

Gruverman[12] reviewed the use of Mössbauer technology in aerospace applications. Broadly, the Mössbauer effect is capable of measurements involving detection of movement at very low velocities; detection and measurement of many elements with unequivocal identification; determination of chemical forms of an element in a sample; determination of changes in temperature, pressure, vibration frequency, and other physical parameters; and tracing unequivocally a stable tracer material. There are two types of applications, depending on whether the absorbed or the scattered low-energy gamma ray is used. One simplification of technique is important: the determination of the Mössbauer velocity spectrum by a calibrated velocity spectrometer is replaced by a library of sources in various matrices providing the pure, narrow Mössbauer lines without complex instrumentation.

Applications to aerospace have not been as numerous as might be expected. Mössbauer analysis could be used to analyze alien specimens nondestructively. It could be used to detect very small departures from true axial rotation of a rotating member such as a high-speed turbine. A Mössbauer source can function as a transducer for many types of physical parameters. In some applications a Mössbauer scatterer could be used as a tracer. Since the Mössbauer effect can monitor environmental changes at the molecular level, it can be used in studies of strain and fatigue in metals.

A technique for measuring the erosive burning rate of solid rocket propellant using a 2.5-curie ^{60}Co source was described by Osborn and Burick.[13] Erosive burning is the change in linear burning rate due to the flow of combustion gases parallel to the receding propellant surface. A severe difficulty in studying this problem is the absence of data on propellants burning under conditions similar to those in rocket motors. A measurement system was devised whereby a propellant sample was burned in a two-dimensional test section; a shaft attached to the back of the sample moved the sample forward at the same speed that it burned back, so that the burning surface was maintained at a fixed position in the test section. A gamma source functioned as a density gage to detect the burning front and feed these data through a servomechanism to move the shaft to keep the burning surface in position. The linear travel of the shaft with time then was a measurement of the burning rate.

Johnson and Murakami[14] described an air-density sensor used in the X-15 research aircraft. The sensor must measure free-stream air density from sea level to an altitude of about 300,000 ft to within ±5%. The sensor must be extremely rugged; temperatures in a typical X-15 flight will vary from −62 to 371°C, and shocks of up to 15 *g*'s are experienced. The air density itself is highly variable, ranging over about six orders of magnitude. The sensor constructed consists of an X-ray tube source, a radiation detector, and associated electronics for data processing. Direct measurement of free-stream air density is accomplished using X-ray scattering.

A gamma-ray spectrometer for space applications is being designed and built, according to Marshall, Weeks, and Halpern.[15] Measurement of the energy spectra emitted by astronomical bodies has been proposed as a method to confirm or to disprove theories concerning stellar formation of heavier elements; the spectrometer described will be used in an earth-orbiting spacecraft for just such an experimental measurement. A flight-worthy prototype is under construction. Preliminary specifications for the final flight instrument call for a highly directional spectrometer with an 800 cm^2 array of 48 detectors, with an energy resolution of 10 kev over an energy range of 50 kev to 2.4 Mev. The detector subsystem will be made up of 48 coaxial lithium-drifted germanium detectors. The engineering approach used in the instrument design and testing gives confidence that the results are realistic. Components requiring precise adjustments because of temperature variations were not used. Preliminary tests indicate that development of a high-precision, ultra-stable gamma-ray spectrometer system is feasible.

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Health and Safety

Blake[16] reviewed nuclear safety requirements in space applications. Future space-exploration efforts will be dependent on the amount of power the space vehicle can carry. Radioisotopic heat sources offer a reliable, long-lived, and lightweight source of power. However, anyone designing a nuclear power supply in a space mission has a moral obligation to provide a system that has been designed for maximum safety. In the United States, development of nuclear power supplies for use in space is regulated by the USAEC, specifically the Space Electric Power Office (SEPO). A guide, "Safety Documentation Requirements for Space Nuclear Power Systems," has been prepared by SEPO for its contractors' use; the guide specifies what documentation is necessary and when it is needed. The preliminary safety analysis is particularly important because it identifies the potential hazards of a particular application. Steps can then be taken to reduce specific risks. The safety of nuclear power supplies is steadily increasing, but tomorrow's systems must be even more safe.

A nuclear safety analysis of a Pm_2O_3 radioisojet thruster was presented by Parker, Pyatt, and West.[17] The radioisotopic heat source is used to expand a gas and provide thrust for attitude control; the radioisojet was successfully hot fired on the ground late in 1966. In virtually all isotope space systems, the most difficult problem is safe return and disposal of the fuel, which must be safely contained in the event of a launch-pad fire, an abort, a short orbit, or after many years at high-temperature operation if the mission is successful. Under these conditions the radioactive material must either be completely burned up and dispersed or remain fully encapsulated even after impact. The most dangerous condition is an abort very late in an ascent, when the impact point cannot be accurately predicted. The analysis of this case for the radioisojet showed that booster abort before 350 sec would allow intact reentry of the encapsulated fuel but that abort after 350 sec would be unsafe because the radioisotope would have its encapsulation breached. Redesign of the radioisojet calls for minor internal modifications plus the addition of a reentry heat shield, with a weight increase of about 40 to 60%.

Anno and Easton[18] assessed the population exposure risk from random reentry of isotopic power devices. The fuel forms used with radioisotopes reduce the hazard greatly, but there is a possibility that the public could be exposed to radiation as a result of reentry. In determining this risk, geographical impact probability for random reentry from a circular orbit, global population distribution, and surface-layer meteorological diffusion stability were considered. Calculations were based on a release of 1 curie of ^{238}Pu and compared to ^{90}Sr. Inhalation doses were used as the exposure mode, and the critical organ of reference was the lungs. Exposure probabilities decline with increasing numbers of exposure or with increasing dose. The probability of exceeding a 15-rem dose from random reentry is on the order of 10^{-3} to 10^{-4}, and this is considered to be a high value.

Odland[19] described the U. S. Air Force (USAF) Radiological Health Laboratory at Wright–Patterson Air Force Base which was developed to support health physicists and bioenvironmental engineers on a worldwide basis when the increased use of radioactive materials and ionizing radiation in aerospace made this necessary. The laboratory operates the film dosimetry service for the USAF, processing about 12,000 films monthly. It has complete facilities for chemical separation, concentration, and purification of radioactive material from biological and environmental samples. Counting facilities are extensive. The laboratory is also the location of the USAF Radioisotope Committee that is the official point of contact between the USAF and the USAEC on matters relating to licenses issued to the Air Force.

Detectors

The use of semi-insulating CdTe and GaAs for gamma detection was evaluated by Zanio, Akutagawa, and Mayer.[20] There is a demand for gamma detectors operating above room temperature, which these materials might fulfill because of their wide bandgaps (1.45 ev) and high effective nuclear charge (50 for CdTe and 32 for GaAs). Response to ionizing radiation is also dependent on the transport properties of materials: drift mobility, mean-free-drift time, and detrapping time. The transient pulse in semi-insulating materials has two components. The fast component is nearly temperature independent, whereas the slow is not. Increasing temperature increases pulse heights. For most applications, pulse-height spectra are desired which are independent of temperature. The transient pulse can be sampled at a point to decrease the temperature effect. Detection of gammas with semi-insulating material is feasible using either the fast component or the total component, the choice depending on the signal-to-noise ratio.

Johnston and coworkers[21] gave the results of testing the high-temperature performance of an ava-

lanche radiation detector. Gamma sensitivity measurements were made of four contoured avalanche detector–tunnel diode instruments, over a temperature range from 30 to 100°C. A detection technique that couples a contoured avalanche detector to a tunnel-diode charge-sensing circuit overcomes some thermal problems because the charge generated by each nuclear event is collected before the time-dependent thermal noise level becomes appreciable. In this study the gamma counting efficiency of each instrument was obtained relative to the incident photon flux measured with a scintillator photomultiplier detection system. A number of radioisotope sources were used as sources of ionizing radiation: ^{55}Fe, ^{125}I, ^{171}Tm, ^{109}Cd, and ^{57}Co. Counting efficiency remained relatively constant when the bias voltage supplies of the contoured avalanche detector and the tunnel diode were adjusted for temperature. Leakage currents severely reduced sensitivity above 100°C.

Trice[22] reported on recent advances in contoured silicon detectors—i.e., silicon p–n junctions in which surface electric fields are limited to values far below that in the interior of the detector. They can operate at 100°C because of their high speed and low noise. The technology of silicon is advanced because of the need for silicon in the communications field, but CdTe offers inherent advantages over silicon. The contoured silicon detector is superior to the standard photomultiplier according to recent measurements of infrared light. The response of contoured silicon detectors to low-energy X rays has been studied. If a soft X ray is stopped in silicon at all, it gives up all its energy in a photoelectron plus usually an Auger electron. Resolution was best at about 5 kev.

Harvey and Bernou[23] described a new development in silicon p–n junction detectors for high-temperature operation. High-speed flight involving high skin temperatures, coupled with the highly desirable accuracy of nucleonic fuel gaging, has produced a demand for a reliable high-temperature solid-state radiation detector. Three design temperatures were considered—70, 120, and 300°C. The first can be handled with existing silicon detectors, and the third will have to be constructed from some as yet unidentified material. The 120°C case was the most interesting. A detector developed earlier, in which the separately connected guard ring was replaced by an unbiased densely doped diffusion ring called a bandguard, was attempted, inasmuch as some units constructed earlier had withstood temperatures of 150°C without bias. This low-yield experimental p–n detector process has been converted into a controlled high-yield manufacturing process. Three changes in the process were necessary. Because the cotton swabs used at one point in the cleaning were found to be contaminated with small quantities of stainless steel (from the combs used in their manufacture), a nitric acid boil was included after the swabbing rather than before; this doubled the average breakdown voltage of diodes in the lot. Because ultrasonic bonds were not produced that could withstand repeated cooling to 77°K (−196°C), a thermal-compression-bonding process was developed that gave satisfactory results. An organic tropicalizing was eliminated when degradation in performance after 150 hr was noted in the detectors.

The original design (Fig. 3) had one p^+ boron-doped ring between the outer edge of the phosphorus-doped disk and the inner edge of the wraparound to

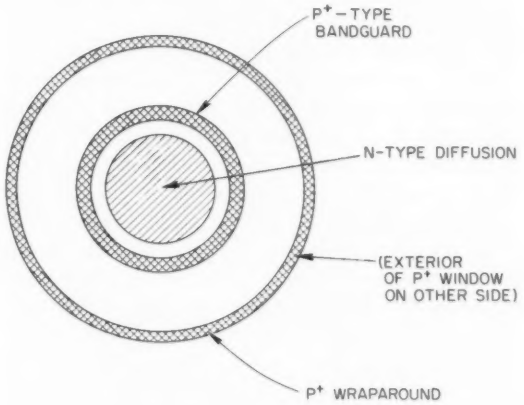

Fig. 3 Basic n-on-p detector with bandguard.

interrupt the n-type surface channel. However, the bandguard must be close to the n-type diffusion to minimize the n-channel extension of the diode's designed area; further, the bandguard effect can be approximated by extending the p-type wraparound to approach the n-type doped disk. The current-reducing ability can be improved by adding a second bandguard. Results showed that 66% of the units produced under the new process were operable at 120°C as far as stability is concerned; the present yield is 43% before on-bias heating, so the overall yield is 28%. Alpha spectroscopy performed with these units indicates that they could be used at 120°C for low rate counting of particles with energies greater than 400 kev.

Bullis and Coleman[24] described a research program now in progress at NBS in which an attempt is made to

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identify the parameters that characterize germanium and silicon of sufficient quality to be used in nuclear radiation detectors. Four basic types of semiconductor radiation detectors are in general use: silicon surface-barrier, silicon lithium-compensated, silicon diffused-junction, and germanium lithium-compensated detectors. Characterization of material as well as characterization of detector afterward are important for development of specifications and test methods used to select detector-grade materials. Material evaluation involves effects of initial-material properties and fabrication procedures on the equilibrium carrier density and the drift-length parameters for holes and electrons. Detector performance is determined by measuring forward and reverse leakage currents, electrical noise, capacitance of junction depletion layers, energy resolution, and charge collection efficiency.

The acceptor density is an important parameter in germanium used in lithium-compensated detectors. This is usually determined by measuring resistivity; however, resistivity by itself tells nothing about compensation. Carrier lifetime, determined by photoconductive decay measurements, is useful but not fully understood; further, it can be affected by deep trapping centers. The presence of oxygen in small amounts is detrimental. (Three methods are used to detect oxygen: infrared absorption at 11.7 μm, lithium precipitation, and lithium drift rate. In the rejected germanium crystals studied so far, no absorption that could be attributed to oxygen has been found, drift rates are nearly the same as those expected in germanium without oxygen, and precipitation results are not available.) The drift mobility can be influenced by other factors than oxygen; an American Society for Testing and Materials test is now being developed to see if "good" material can be detected by some easy standard method. The method is intended for use on geometrically regular slices of *p*-type single-crystal germanium.

Carrier mobility, dependent upon both electrons and holes, is sensitive to ionized impurity density, especially at low temperatures. Hall measurements have been used to test for impurity density but with poor results. Photoconductive decay lifetime measurements and reverse recovery time measurements have also been attempted; their significance is unclear because the necessary relationships have not been defined. After fabrication into a detector, the tested germanium material is further evaluated. The forward and reverse current–voltage characteristics and the capacitance–voltage characteristic are measured with the detector at the operating temperature of 77°K (−196°C). The forward current–voltage characteristic indicates the quality of the contacts that have been applied or diffused on a detector. In a lithium-compensated detector, the capacitance–voltage characteristic can be used to measure the profile of the net charge distribution of the drifted depletion region. The noise–voltage characteristic is often an indication of the quality of the final junction surface treatment. Noise measurements of the detector and the electronic system can be subtracted from the observed energy resolution to evaluate charge transport problems that exist in a detector. An estimate of the magnitude of the mobility-lifetime product can be obtained by a measurement of the collection efficiency as a function of the electric-field strength. An even more sensitive estimate of the mobility-lifetime product can be obtained, in principle, from a study of energy resolution as a function of the detector electric-field strength.

A paper[25] prepared by the technical staff of Metallurgie Hoboken, Belgium, argued that lithium mobility is not a sufficient criterion to characterize germanium for suitability for detectors, but that lithium precipitation is an important property. When a crystal does have a high mobility, precipitation should be low so that rediffusion is not required. Precipitation rate is evidently dependent on diffusion conditions; temperature during the 150 hr of drifting is more important than the time. High mobility generally corresponds to low precipitation rates as long as quenching and surface state are of high quality.

Weisberg and Goldstein[26] reported on the development of GaAs and GaP for room-temperature gamma-ray counters. Only silicon has been suitable for room-temperature gamma-ray detection in the past because other materials suffered from excessive trapping. The GaAs–GaP system can tolerate a large trap density without necessarily degrading the collection efficiency because it has a higher atomic number, higher mobility, and higher resistivity than does silicon. Further, GaAs has a larger ratio of electron-to-hole mobility (>10) than does silicon. The desired properties of these materials are not reproducible; silicon resistivity of 10^4 ohm-cm at room temperature is more difficult to achieve than the resistivity of 10^8 ohm-cm achieved in GaAs by doping with chromium or iron. The state of the art of making contacts—either blocking or ohmic—to GaAs is quite well advanced, and vapor-phase growth has been used to reduce problems of homogeneity, segregation, and facet effects. Many more traps are allowable in GaAs materials because in GaAs the mobility is higher, the thickness

can be smaller, and the applied field can be larger. For a counting efficiency of 0.9, the concentration of trapping centers can be as high as 10^{15} per cm^3; for a higher efficiency the concentration of trapping centers will have to be lower. Four advances in material preparation are (1) introduction of arsenic and phosphorus via their volatile species to give increased homogeneity; (2) increasing growth rate of the vapor growth method to 3 μ/min; (3) doping with iron or chromium during the vapor growth; and (4) achieving less trapping than ever before because of the high purity of the vapor growth method. The high resistivity of both silicon and GaAs detectors is attained by the inclusion of impurities that provide relatively deep energy levels; the advantage of GaAs is that the impurities provide a greater depth of the energy level, and thus the GaAs material can be used in a detector at room temperature.

Heat Sources

Perkins and Hedgepeth[27] described a method of infrared detection of radioisotope heat sources. An airborne system of some kind would be necessary to locate the source after a mission abort or reentry. A large radioisotope heat source was simulated by a 40- by 48-in. stainless-steel plate; a small radioisotope heat source (or a fuel capsule from a nonintact reentry) was simulated by a 12-in. length of 1-in. stainless-steel pipe. Both test objects were heated electrically. An infrared device that displayed real time variations for temperature changes of 2°C was flown in a twin-engine aircraft at altitudes varying from 2500 to 10,000 ft over the heat sources. The sources were placed in the open, buried under 2 in. of sand, or placed under heavy foliage. For the open test the plate was maintained at 496°C and the capsule at 427°C; they were positively located on the first pass at 2500-ft altitude. However, at 5000 ft they could not be separately identified. The sources were concealed under heavy foliage, with the plate at 507°C and the capsule at 749°C. Because of heavy cloud cover, the heat sources were not located until a fifth overflight at 3500 ft; the diesel engine powering the electrical generator was also located (exhaust stack temperature was 227°C). The large heat source was buried under 2 in. of sand for the final test. The heater temperatures became excessively high, and the plate was not maintained at a constant temperature. The plate temperature varied from 982°C down to 478°C; the sand surface temperature varied from 182°C to 161°C. From altitudes of 1500 to 2500 ft, there was no difficulty in locating the buried heat source.

Target sources were identifiable at altitudes up to 10,000 ft in clear weather; in fact, at the high altitudes the sources were more easily identifiable because the background was greatly diminished. However, the target sources could not be located through clouds, hence probably not through rain. The discrimination of the detecting system could be improved by changing from an imaging system to a point-source detection system or to a combination of the two systems since identification of the background heat sources would aid in specifically locating the target heat sources.

Shivers[28] reported the development of a radioisotope swimsuit heater (Fig. 4). The temperature in

Fig. 4 Swimsuit heated with radioisotopic energy.

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deep water on the continental shelf reaches 5°C and imposes a thermal limit on underwater exploration. Survival at low temperatures is also a problem faced by astronauts, aviators, and arctic and antarctic explorers; these men would also benefit from development of a suit heater. The specifications arrived at for the suit were regulatable heat, with a maximum value of 270 watts(t); low radiation, with usage allowed at least 5 hr/day for 30 days/year; usable in water at 7°C; and maximum weight of 15 lb. Later the specifications were changed to heat of 400 watts(t) and a weight less than 20 lb. The heat source used was the SNAP-19C capsule, which is based on ^{238}Pu. The heat source and heat exchanger must not interfere with the normal duties of the swimmer, of course. The unit has been fabricated but not yet incorporated into a swimsuit. Tests show that the unit will provide 390 watts(t) to the suit with water flowing at 1.6 to 1.8 liters/min. The capsules will withstand about 33,400 psi internal and 27,300 psi external.

Another use of a radioisotope to provide heat by its own nuclear decay was presented by Levy and Bustard.[29] The performance of Ag–Zn batteries is inferior in a low-temperature environment; a heat source using depleted ^{238}Pu (20 to 40%) was selected to maintain the batteries at an optimum temperature. The capsule assembly for the heater consists of a simple box enclosing six cylinders. This box will fit between the two cells of the battery (Fig. 5) for optimum heat distribution; the cells will also function partly as safety shields against radiation. The raw-fuel cost will be about $1450 per heater, but the heater can be reused for many batteries. The cost per battery is thus reasonable. The battery will be kept at an operating temperature of 4°C, and the isotope heater will be used whenever the ambient temperature drops below −1°C.

Metzger and coworkers[30] described the application of radioisotopes to aerospace waste-reclamation and water systems. The economics of long-term space missions dictate the recovery of water for drinking purposes from feces, urine, perspiration, and exhaled vapor of the astronauts. The system developed must be reliable and must meet stringent aerospace specifications, especially that of operating under zero-gravity conditions. Radioisotope heat sources are particularly suited to use with vacuum distillation–vapor pyrolysis systems, and a system of this type was evaluated and fabricated. Design requirements were recovery of 90% of the water in 15 lb of urine daily for 30 days under the environmental conditions of normal and zero gravity, a temperature range of 15.6°C, pressure from 380 to 760 mm Hg, humidity from 30 to 80% (relative), and atmosphere both normal and 100% oxygen.

The waste liquid is metered into the evaporator where it is heated to 48.9°C by a 210-watt radioisotope heat source at a pressure of 87.5 mm Hg. The

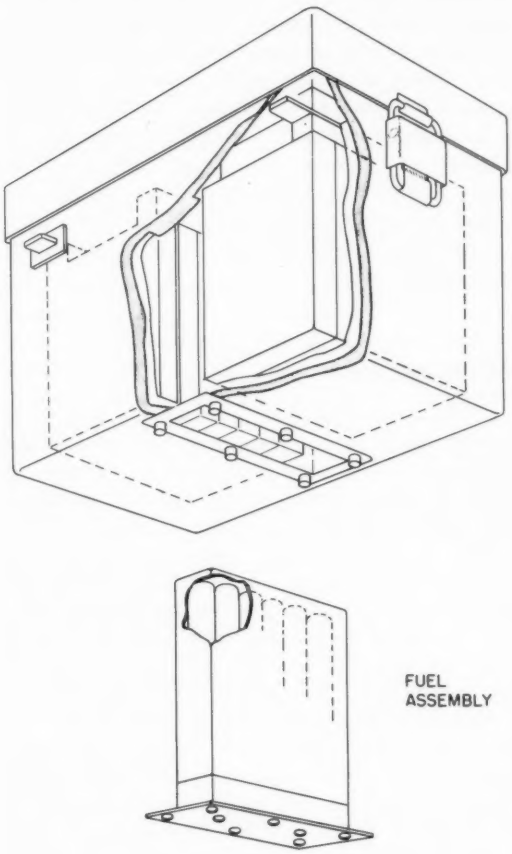

Fig. 5 Radioisotope-fueled Ag–Zn battery.

vapor is passed through a superheating heat exchanger (to ensure that no liquid is carried with the vapor flow) to a pyrolysis unit where it is heated to 650°C in the presence of a ruthenium or rhodium catalyst. The heat is provided by a 70-watt radioisotope heat source. The impurities are removed via the vacuum vent in the condenser. The vapor is condensed at 15.6°C, which establishes a pressure of 13.2 mm Hg; this pressure differential between the evaporator and the condenser is the driving force that moves the vapor through the

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system. Solid wastes removed from the liquid are centrifugally conveyed to the storage reservoir by the impeller in the evaporator. The hot-water reservoir is filled before the cold-water reservoir, and the hot-water reservoir is heated by the steam from the pyrolysis unit.

A second unit that was designed, developed, and fabricated was a vacuum distillation–vapor filtered system. This system has few moving parts. As in the previous system, a radioisotope heat source supplies the thermal energy to evaporate the waste liquid. The vapor passes through a microporous membrane and a catalytic oxidation unit to the condenser.

Nondestructive Testing

An aircraft-engine inspection technique using a radioisotope as the radiography source, with a well-collimated beam, was reported by Packer, McKenzie, and Bruton.[31] The basic technique of applying isotope radiography to gas-turbine engine inspection consists of positioning a radioactive source within the hollow engine shaft and exposing film on the exterior surface of the engine; however, considerable secondary gamma rays are produced by Compton scattering when using this technique. Lead-film filters have been used to screen out low-energy radiation, but the most effective means of reduction is selective shielding of the source. A spool-shaped collimator has been developed to attenuate the emitted gamma radiation in all directions except a cone, with the apex at the gamma source and the base at the film. The interchangeable spool pieces provide 360° fields of various widths, single beams at 90° to the center line, or any other desired beam projection. Source collimation not only improves the general quality of a radiograph, it also gives image resolution hitherto unobtainable. Engine regions that were formerly inaccessible can now be inspected. The source can be safely positioned outside the engine for special applications.

Frasca, Bemisderfer, and Krause[32] presented work on using electron backscatter to detect corrosion. A ^{90}Sr–^{90}Y beta source was used. The number and energy of backscattered electrons are dependent on the atomic number of the material. For bare aluminum the average Z is 14; for corroded aluminum the Z drops to about 10.5. The magnitude of the signal change should be about 25%, going from pure metal to the oxide. The gage is designed to scan through an epoxy paint coating and can measure corrosion depths up to 20 mils and as small as 2 mils. The source travels a 4.5-in.-length oscillatory path between two fixed scintillators. This arrangement keeps the scintillator–phototube combinations rigid and reduces the complexity of the design. The source is driven by a cylindrical cam with a small pawl as the indexing component. Calibration corrosion samples were made from a combination of epoxy paint and aluminum oxide to give uniformly shaped corrosion pits so that an accurate determination of the gage sensitivity could be obtained. Results of tests showed that an appreciable amount of corrosion material should be present for the gage to detect surface corrosion; this condition is not necessary for pitting occurring on the underside. The area of the corrosion pit should be larger than the area of the collimating beam port for maximum sensitivity.

The potential for the application of Kryptonates in aerospace was assessed by Figueroa.[33] The use of gaseous ^{85}Kr is widespread in aerospace; oil gaging, landing guidance systems, and helicopter formation-keeping systems are but a few applications. The incorporation of ^{85}Kr into the lattice structure of materials, carried out by ion bombardment or high-temperature diffusion, provides a unique tool for analysis. These Kryptonates are extremely stable at room temperature. Any process that disturbs the bonding results in a change of activity, which makes Kryptonates good indicators of physical processes not detectable by other means. There is a fractional loss of activity when the material is heated; thus Kryptonates have been widely used for surface-temperature measurement.

Kryptonates can also be used to measure fatigue since the behavior of ^{85}Kr in the host material is dependent on the state of the lattice which is affected by the fatigue process. Two methods have been developed, based on the fact that the amount of ^{85}Kr accepted or released by the host material is a function of the crystalline structure and its condition. A material that has fatigue damage will absorb more ^{85}Kr than a nonfatigued material; a kryptonated material will lose some activity as a result of fatigue. A third method uses the difference in ^{85}Kr release shown by fatigued and nonfatigued material at elevated temperatures. Experiments with aluminum stock showed that nonfatigued material retained 8 to 10% of its activity after heating to 260°C for 30 min; in about one-third of the samples of fatigued material, a higher percentage of ^{85}Kr was retained.

Martin and Tsang[34] reported on the effect of space charges on the high temperature fatigue of nickel. A space charge is an unbalanced electric field existing from the interior of a semi-infinite solid across a multilayer interface to a semi-infinite fluid, e.g., air.

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The rate of many surface reactions is controlled by the rate of diffusion of the reacting species; a space charge will affect ion movement and hence the reaction rate. This experiment demonstrated that the effect on reaction rate will also affect surface-sensitive characteristics of a metal, such as fatigue. Nickel was tested at 816°C with fatigue induced by torsion; the space charge was generated by a potential of 1000 volts/cm and external ionization from a 25-kilocurie ^{60}Co source. The fatigue life with maximum ionization in a negative electric field was about 25% longer than in a positive electric field. Original plans called for a 3-curie source using alpha-emitting ^{241}Am; however, the supplier found that he could not fabricate the required source because of leakage problems at high temperature. A more complete test is planned for other materials.

The practical applications of neutron radiography in nondestructive visual examination of aerospace hardware were reviewed by Meyer and Porter.[35] A neutron radiograph is made by exposing an object to a well-collimated beam of thermal neutrons; the beam passes through the object and impinges on a converter screen, carrying in its spatial intensity modulation information about the internal structure of the object. The converter screen—a thin sheet of gadolinium metal—captures the neutrons and emits low-energy X rays that expose a standard X-ray film. The quality of the image is dependent on the quality and intensity of the neutron beam. At present only a well-collimated beam of thermal neutrons from a nuclear reactor is suitable. Materials appear lighter or darker on the film depending on their ability to absorb or scatter thermal neutrons; this ability is an almost random function and is almost completely independent of atomic number. Explosive devices, used widely in the aerospace field, are particularly good subjects for neutron radiography because explosives and epoxy materials are often of hydrogenous material. X rays cannot provide post-assembly examination of many of these devices; neutrons can. Hydriding of titanium welded assemblies is also detectable with neutron radiography.

Polishuk and coworkers[36] covered recent applications of nuclear methods and techniques to aerodynamic testing. Among the techniques used have been activation analysis, kryptonation, wear studies (i.e., ablation), and position studies (using well-collimated sources and detectors). Nucleonic instruments, such as the multichannel pulse-height analyzer, are of use in nonnuclear applications. Specific cases included the use of activation analysis to determine the presence of copper and tungsten particles on an air filter from the high Mach number Gas Dynamic Facility at Wright–Patterson Air Force Base. A potential application is the use of radioisotope-generated plasmas for studying and simulating the transport properties of gases. The environmental density around an aerospace vehicle can be determined by gamma backscattering techniques; variation in local density as the vehicle body departs from a symmetrical alignment with the environmental medium can be used to indicate the angle of attack. A multichannel analyzer was recently used to record the number of times an airfoil tip intercepted a light beam that impinged on a photocell. The photocell output was viewed on an oscilloscope; a trigger output pulse from the oscilloscope then drove a monostable multivibrator that produced a pulse acceptable to the multichannel analyzer. By this means the time a rotating airfoil took to decelerate could be measured accurately.

Sources

Posey[37] reported on an analysis of the consequences of internal heat flow in radioisotope heat sources. Large temperature differences can exist between the surface and the interior of the radioactive fuel (radioactive material with or without a diluent). The melting point of the fuel is sometimes set at the maximum allowable internal temperature, but in some applications melting in the interior of the material does no harm. The maximum allowable internal temperature does limit the maximum surface heat flux and the size of some sources. Equations relating the maximum internal temperatures of various-shaped fuel forms and other source parameters are given. The rate of heat generation per unit volume of the fuel and the thermal conductivity of the fuel are important parameters, and if the product of these two factors can be increased, performance will be enhanced. For material with poor thermal conductivity, the addition of a metal—either by producing a cermet or by introducing oriented conductors—will improve heat conduction. Limited data show that certain radioactive fuels may be volatile under the proposed conditions of use. This volatility causes no serious problems with regard to heat flow; in fact, the radioactive material will become distributed within the capsule in an ideal manner as a result of its volatility. However, a high internal temperature has three major disadvantages: (1) the pressure of the internally stored helium gas increases, (2) the metal wall of the capsule weakens and deteriorates faster, and (3) the heat loss through the insulation becomes greater. An ideal capsule for a volatile radioactive

material where the internal temperature is a limiting factor should have thick walls of a highly conducting metal and should use internal metallic heat paths to conduct the heat to the desired surface.

Ketchen and McHenry[38] reported measurements on the thermal conductivities of SrO, Sr_2TiO_4, and $SrTiO_3$ (or compositions that simulate these compounds prepared from nuclear fission wastes). Thermal-conductivity data on radioactive heat-source materials are necessary for the calculation of thermal gradients in heat sources. Conventional methods require large well-instrumented samples, but the unavailability of sufficient heat-source materials and the difficulty of fabricating instrumented samples render conventional methods impossible. The apparatus used to measure the thermal conductivity is a thermal comparator in which a high heat flux supplied by a heater flows in sequence through a specimen of known thermal conductivity, a specimen of unknown thermal conductivity, and another specimen of known thermal conductivity to a heat sink. The standards used in this work were Pyroceram 9606 cylinders, 1.75 in. long and 1 in. in diameter. The sample was 0.5 in. long and 1 in. in diameter. The sample compound was prepared by adding an amount of TiO_2 to a slurry of strontium and alkaline earths from process waste of fission-product handling. The alkaline earths were precipitated as carbonates and the mixture was calcined at 1200°C to decompose the carbonates to the oxides and, at the same time, to react the oxides with the TiO_2 to form the titanates. No thermocouples were inserted in the sample specimens. Thermal conductivity was measured for SrO from 405 to 747°C, for Sr_2TiO_4 from 359 to 728°C and from 384 to 749°C, and for $SrTiO_3$ from 394 to 813°C and from 408 to 861°C. A function was fitted by least mean squares to the data under the assumption that thermal conductivity is proportional to the reciprocal of the absolute temperature. For SrO, $k = 0.0135 + 7.415/T(°K)$; for Sr_2TiO_4, $k = 0.0261 + 4.449/T(°K)$; and for $SrTiO_3$, $k = 0.0185 + 15.592/T(°K)$. The thermal conductivity of TiO_2 was measured to evaluate the systematic error; it agreed well with NBS data. Systematic error probably does not greatly exceed the precision.

For any given source, specific activity varies with isotopic composition and yields unique X-ray-to-alpha-particle and gamma-ray-to-alpha-particle ratios. These ratios were measured for plutonium by Swinth.[39] Interpretation of X-ray or gamma-ray counts for plutonium samples is difficult because ^{239}Pu, the major constituent of most plutonium samples, has a very low specific activity and X-ray yield so that the number of X rays per alpha particle is sensitive to changes in the presence of other isotopes. For the measurement of the X/α ratio, a proportional counter–scintillation counter combination was used; sources electrodeposited or dried on stainless-steel disks were used. The output of the two counters is fed to a coincidence-gated multichannel analyzer, which is necessary for the accurate intercomparison of low-level sources. Four sources of known isotopic composition were measured, and five sources of unknown composition were measured. Results are given in Tables 1 and 2. A solid-state system has been constructed to improve on the measurements. A lithium-drifted silicon detector is used for the X rays; alpha particles are detected by a surface-barrier diode. Efficiencies are low, although extra resolving power is obtained with this solid-state system. The easily measured X/α ratio yields information useful in evaluating the relative health hazards of plutonium.

Table 1 Measurement of X/α Ratio for Known Sources

Source	Major isotope	X-rays/alpha	Gamma-rays/alpha
1	^{238}Pu	0.112 ± 0.001	5×10^{-4}
2	^{239}Pu	0.0485 ± 0.0004	6.03×10^{-4}
3	^{239}Pu	0.0737 ± 0.0007	0.0191 ± 0.0006
4	^{240}Pu	0.109 ± 0.0005	$(2.32 \pm 0.05) \times 10^{-3}$

Table 2 Measurement of X/α Ratio for Unknown Sources

Source	Alpha activity, dis/min	X-rays/alpha	Gamma-rays/alpha
1	1260	0.0783	0.0197
2	608	0.0768	0.0259
3	1260	0.0770	0.0225
4	292	0.0606	0.0096
5	4.5×10^4	0.0730	0.0108

Mather[40] discussed the dense-plasma focus device as a potential intense source of neutrons. The device takes the general form of a hydromagnetic coaxial plasma accelerator in which the hot dense deuterium plasma is created by the rapid radial collapse or pinching action of a high current axisymmetric current sheath toward the axis as the sheath leaves the end of the center electrode. The neutron production rate per unit volume from a thermalized deuterium plasma

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depends on the square of the particle density and very strongly on the plasma ion temperature. The neutron source is small; it has a diameter of about 1 mm and a volume of about 3 to 5 mm^3. The full-width half-maximum neutron-pulse width is 0.1 μsec; maximum neutron production of 3.5×10^{10} neutrons per pulse has been detected, corresponding to a rate of 3.5×10^{17} n/sec of 2.45 Mev neutrons. With appropriate modifications to the gas used and use of higher energy, the system can be expected to produce 10^{18} n/sec. The dense-plasma focus device cannot compete with reactors as a continuous neutron source, but as a pulsed source of monoenergetic neutrons, the only device that surpasses it is a nuclear explosion.

August[41] gave the results of an investigation of ionization of air by alpha particles to enhance the electrical conductivity of the gas. Primary ionization is caused by collisions between alpha particles and gas particles, and many of these collisions produce sufficiently energetic electrons to cause secondary ionization. Two sources were used: a 5-in.-diameter disk coated with 30 mc/sq in. of ^{210}Po and a 0.25-in.-diameter source coated with 1.9 curies/sq in. of ^{147}Pm. A microwave technique was used to measure the plasma induced in air by the alpha radiation. The plasma properties are indicated by the degree of interaction that occurs between the beam and the plasma. Results indicate that air at normal temperature may be induced through alpha radiation to attain an electrical conductivity equivalent to that of a thermal air plasma at approximately 2500°K.

Results of recent experiments using a flash neutron generator were reported by Hollister.[42] The generator is a focused plasma device that generates neutrons by a (d,t) reaction within the plasma rather than by accelerating deuterons into a tritium target. The generator has been operating about a year; it provides 10^{11} n/pulse, and each pulse is 10 to 100 nsec long. It has been used to induce ionization in semiconductor devices. Comparisons with semiconductor devices ionized by photons from a flash X-ray machine gave a dose equivalence of 1.24×10^{-9} rad/n; the number commonly accepted for irradiation of bulk semiconductors is 0.86×10^{-9} rad/n. The flash neutron generator was also used to determine the fluorescence decay time under neutron irradiation of the fluorescent materials calcium tungstate, zinc sulfide, and thallium-activated sodium iodide. The decay time was found to be essentially the same as under X-ray irradiation. Promising applications that are being investigated are calibration of low-sensitivity neutron detectors, short half-life activation analysis, and neutron radiography.

Analytical Techniques

The surface distribution of carbon and oxygen was measured by ^{3}He activation and autoradiography according to Holm and coworkers.[43] The amount and distribution of oxygen and carbon in metals affect their physical properties. Activation analysis by the reactions $^{16}O(^{3}He,p)^{18}F$ and $^{12}C(^{3}He,^{4}He)^{11}C$ provides a sensitive method for measuring these impurities. Both activation products are positron emitters; their half-lives are 110 min for ^{18}F and 20.4 min for ^{11}C. The decay history is followed by a pair of NaI detectors in coincidence, and a series of autoradiographs is made concurrently. Samples of Ge, Ta, Zr, steel, Ni, and stainless steel have been irradiated and measured. Segregation of oxygen and carbon has been observed along grain boundaries. By suitable etching the distribution under the original surface can be measured and surface contamination with oxygen and carbon eliminated.

Lukens and Guinn[44] described a neutron activation analysis method for the determination of very small amounts of lithium in various materials. A reactor was used to effect the $^{7}Li(n,\gamma)^{8}Li$ reaction. The short life of the energetic beta (0.84 sec, 13.1 Mev) emitted by the activation product necessitated use of a rapid pneumatic transfer tube between the reactor core and a well-type Cerenkov counter. Because ^{8}Li can also be produced by the $^{11}B(n,\alpha)^{8}Li$ reaction, samples of both boron and lithium were irradiated and counted. Counting rates observed were 1.0×10^{3} counts per sec/μg of Li and 1.28 counts per sec/μg of B; sufficient discrimination was thus demonstrated. Pulsing the reactor gave a detection rate of 1.38×10^{5} counts per sec/μg of Li, allowing detection and measurement of as little as 0.001 μg of the element.

Activation analysis was used by Andersen[45] to determine oxygen in titanium and refractory metals. A rapid method is needed for the aerospace industry inasmuch as the new refractory metals are being used increasingly, but no routine and reliable chemical determination method is yet available. The $^{16}O(n,p)^{16}N$ reaction, employing fast neutrons, is best suited for the average analytical installation. The system used consists of a $^{3}H(d,n)^{4}He$ neutron generator, a fast pneumatic sample-transfer tube, and two 5-in. NaI(Tl) scintillation crystals, with the outputs summed and fed into a single channel pulse-height analyzer. The standard used was an intimate mixture of oxalic acid and spectroscopic grade graphite in a polyethylene container, taking into account the oxygen in the graphite and the container. The fast-neutron

activation analysis method was compared to the vacuum-fusion method and the inert-gas-fusion method; results were comparable. The advantage of the neutron activation analysis method is its speed.

Jones and Carpenter[46] reported the sensitivity of a nondispersive X-ray fluorescence spectrometer for multielement trace analysis. Semiconductor detectors have lacked the necessary resolution for X-ray fluorescence analysis. However, semiconductor detectors have several advantages: response is proportional to energy of the impinging photon, response to radiation is very stable, no high voltage is necessary, and stopping power is high compared to proportional counters. The device described here uses cooled-field-effect transistors in conjunction with lithium-drifted silicon crystals to provide a detection system with resolution superior to 350 ev full-wave half-maximum (FWHM). A radioactive source (^{125}I) irradiates a sample that absorbs the radiation and then either reemits characteristic X rays or scatters the primary radiation. The detector responds to the radiation by generating current pulses proportional to the energy of the radiation; these pulses are converted into voltage pulses and fed into a pulse-height analyzer. Liquid samples were used to simplify standardization, provide uniform elemental distribution, give easy control of concentration, and eliminate particle-size effects. The ^{125}I was actually five 1-mc sources in an annular ring. Elements tested were Mo (2 ppm to 0.05%), Sr (5 ppm to 5%), Rb (10 ppm to 5%), and Br (10 ppm to 5%). The assumption that counting rate varies linearly with concentration was found to be valid for concentrations up to 1%; above 1%, self-absorption affected the results. Some problems associated with applying this spectrometer to field work are providing cooling to liquid-nitrogen temperatures (in aerospace applications, ambient temperature would be adequate for operation), making the equipment rugged enough to withstand vibration and shock, and accomplishing remote calibration and checkout.

Nelson and coworkers[47] gave results of determining fast-to-thermal-neutron ratios by Ge(Li) gamma spectrometry. The neutron source used was a Cockcroft–Walton accelerator that produces neutrons through the reactions $^{3}H(d,n)^{4}He$ and $^{2}H(d,n)^{3}He$. Neutrons are usually divided into three classes: (1) thermal neutrons ($E < 0.4$ ev), (2) resonance neutrons (0.4 ev $< E <$ 100 kev), and (3) fast neutrons ($E >$ 100 kev). Foil activation is the usual technique used for neutron-flux measurements, with indium, gold, and dysprosium being the three classical thermal-neutron-activation foils. Because of the distance between the generator and the laboratory where the foils were counted, gold foils were used since gold has the longest half-life of the three foils. Two reactions in gold are of interest—$^{197}Au(n,\gamma)^{198}Au$ and $^{197}Au(n,2n)^{196}Au$. The first is a thermal-neutron reaction and the second a fast-neutron reaction (threshold energy = 8.11 Mev). A cadmium shield was used to block resonance neutrons. Foils 1.13 cm in diameter and 0.05 cm thick were exposed in the neutron generator room, one covered with cadmium and the other bare. Measurement of the 0.412-Mev gamma from ^{198}Au and the difference between the two foils gave the thermal-neutron flux. Measurement of the 0.356-Mev gamma from ^{196}Au would give the fast neutron flux, but yields were too low. Instead, a separate measurement was made using the $^{63}Cu(n,2n)^{62}Cu$ reaction. The results showed that the thermal-neutron flux around the neutron generator is very slight.

The possibility of using radioisotopes as zero power sources of X rays for X-ray diffraction analysis were covered by Toothacker and Preuss.[48] If a radioisotope source can be substituted for the X-ray tube structures now planned, there would be a substantial savings in weight and space aboard any aerospace vehicle inasmuch as no power supply would be needed for the radioisotope source. The main problem of such a system is the relatively low intensity generally available from radioisotope X-ray sources. This leads to another problem, that of resolution. Iron-55 was chosen for study. This isotope can be produced in a reactor, but the product is usually contaminated (for the purpose of this experiment) with ^{59}Fe and ^{59}Mn. An accelerator can produce ^{55}Fe of sufficient purity. A source of 100 mc was prepared at Oak Ridge National Laboratory with a shape and dimensions (2 x 10 mm) approximating those of a standard X-ray tube diffractometer. Self-absorption and uneven distribution of the radioactive material caused some not insurmountable problems. Testing a powder sample of LiF, the system gave a resolution of 0.3° FWHM, which does not quite meet the requirement of 0.25° resolution. In a lunar application the intensity of the source would be nearly doubled because of the absence of air absorption. Slow scanning speeds and multiple detectors might also be used to make use of a radioisotope source of X rays feasible.

Ranging and Guidance

Petriken[49] described the requirements of a system that would enable the Army to fly close helicopter formations in marginal weather and at night. Such a

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system would increase air mobility, reduce the probability of enemy detection, allow groups of aircraft to take off and land faster, reduce vulnerability of aircraft by decreasing volume of airspace occupied, and increase safety of air operations. Since Army helicopters—such as the UH-1B, CH-47, and UH-1D—are neither spacious nor expensive when compared to other support aircraft, the formation system must be compact, light, and inexpensive. There is little room in a helicopter cockpit for complex display screen. The system must determine the relative positions in three dimensions of all aircraft in the formation, including measurement of range, range rate, bearing, relative elevation, and angle rate. It must be compatible with the airframe and must provide timely data to the pilot. It must also provide a proximity warning capability to prevent collision. The sensor must have sufficient range not only to guide the helicopters in the flight but also to make possible rendezvous with armed escort aircraft. Another essential requirement is capability of rejection of false signals or jamming by an enemy.

Army studies have concluded that formation flight should continue to be based on a platoon concept. Formation flight should be restricted to a particular volume of airspace. It increases safety, reduces air-traffic-control complexity, and maintains the concept of air mobility. Formation flight is feasible at short ranges (75 to 1000 ft) by use of nucleonic techniques. The University of Pennsylvania is currently developing a mathematical model and computer simulation of the behavior of a company of helicopters. Princeton University is studying the influence of aircraft dynamics and control characteristics on formation-flight accuracies under gust conditions at all speeds in an automatic mode. Minneapolis-Honeywell Regulator Co. is undertaking to define the parameters of a display system.

A test facility to be used to evaluate problems of formation flight was described by Rubinstein and coworkers.[50] The facility will consist of a 2000-ft-long test range 300 to 400 ft wide. Every 100 ft there will be a data-acquisition station. There will also be a mobile X-ray transmission station, a mobile data-acquisition center, a mobile detection van, and an X-ray transmission tower, which is also used as an observation tower for the range. X rays are generated either by the pulser in the tower or the pulser in the van to permit measurements at varying ranges and bearings.

Rubinstein[51] reported on a formation-flight system using high-quantum-energy radiation. High-energy rays include radioisotope-emitted X and gamma rays and tube-generated X rays. Some of the advantages of using high-energy radiation are: (1) all-weather operational capability, (2) high short-range accuracy, (3) immunity to enemy countermeasures, (4) excellent security—the noise from the engine will give away the position before the ranging system does, (5) light weight, (6) already developed and available detectors, (7) elimination of multipath problems—especially with pulsed X-ray sources, (8) conservation of radio-frequency spectrum that is already overcrowded, (9) low relative cost, (10) no scanning required, (11) no external modification of aircraft, (12) low volume, (13) applicability to entire line of military aircraft, (14) low-altitude capability, and (15) multifunction-sensor possibility. The source of radiation chosen for most consideration is the pulsed tube-generated X ray. It provides a high dose rate that permits increased range and safety; it has reduced power and size requirements; its use eliminates multipath problems. Simple modulation leads to pulse coding, and gating techniques can prevent dose buildup. In addition, background noise can be rejected, and a tube-generated X ray is more readily adaptable to multifunction-sensor capabilities. Range can be measured by measuring either the amplitude of the detected signal or time of flight between pulse initiation and detection. Pulsed amplitude techniques do not require identical systems on all aircraft; follower aircraft need carry only detection systems. Using a number of pulses and integrating at the detector will permit range and angle measurements with increased reliability at greater distances. A disadvantage of pulsed X-ray operation is that the distribution of radiation is not omnidirectional; the information provided to detectors is low in intensity if they are off-axis. Detector-system instabilities pose some problems, but a small isotope source can be used in a constant-circuit gain to stabilize the output of the detector system. The photon count for a time-of-flight measurement is much lower than the photon count for an amplitude measurement, but angle measurements do require an amplitude measurement. Using relative amplitude to provide information on bearing is difficult at long range because the source appears "smeared." Ranging determination must occur often enough so that no significant changes occur between determinations; five determinations per second are adequate, and five pulses per second are easily obtainable.

The basic design goals for a system weighing 30 lb and operational at altitudes up to 20,000 ft are: range, 75 to 2000 ft; range accuracy, ±10%; bearing coverage,

360°; relative altitude coverage, ±45°; and relative altitude accuracy, ±10 ft in 75- to 500-ft range and ±20 ft in 500- to 2000-ft range. The pulsed X-ray time-of-flight technique appears to offer the most promise as a high-energy radiation system to solve formation-flight problems, and the Army is continuing to investigate this and other high-energy approaches to the problem.

A pulsed coincidence X-ray system for formation flight was described by Feinstein.[52] A mathematical model that recognizes the spherical nature of waves originating at a point and uses the changing magnitude of the field of curvature to determine range was derived. A large error factor in an X-ray system may be tube and transponder jitter. A technique is needed that uses only one-way transmission, eliminates the need for a source of known magnitude, does not use detector shielding to determine bearing, and eliminates the error derived from tube jitter. The technique derived from the mathematical theory led to placing one detector in a fixed position and a second detector on the end of a 25-ft rod rotating about the fixed detector. The delay intervals between detection can be directly correlated with the curvature of the field emitted by the radiation source, giving very accurate results at close range.

Johnson[53] covered the proposed application of X-ray ranging to measurement of lunar altitude and altitude rate. A pulsed X-ray system would provide greater low-altitude accuracy than radar would; it would not be affected by lunar dust or ionized rocket exhaust and would not require antennas or windows. The X-ray machine was chosen over a radioisotope because of shielding and safety problems. The two-way pulse travel time is converted to an analog voltage and displayed to the astronaut. The system has been assembled and will be tested soon.

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X Rays from Radioactive Isotopic Sources for Diffraction Analysis*

By Luther E. Preuss and William S. Toothacker†

Abstract: *The use of X rays from beta-excited and electron-capture X-ray sources is reviewed critically. The difficulty caused by self-absorption of soft photons because of the geometry requirements is discussed. Sufficient intensity will probably not be achieved for practical use with beta-excited sources, but electron-capture sources such as ^{55}Fe have definite promise, especially for portable use in remote locales.*

In recent years, X rays originating from radioactive sources have been applied to various analytical problems for which X rays from a standard X-ray tube had been used previously. Notable advances in nondispersive analysis have been achieved by the use of isotopic X-ray sources. A natural step would be to evaluate, also, such X rays for dispersive analysis and thus to take advantage of the considerable benefits of the isotopic source. A project of this type has been treated by our laboratory over the past few years, and two different sources have been studied in a series of experiments. The first involved the electron-capture isotope ^{55}Fe, which produces the characteristic K X-ray emission of manganese. The other was a ^{147}Pm–Cu apposition source, which produces rather efficiently the K X rays of copper, plus a bremsstrahlung continuum, by the impingement of beta rays from ^{147}Pm upon the copper target.

*The support of the USAEC, Division of Isotopes Development, is herewith acknowledged.

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Preliminary Results

An X-ray source of a radioactive material for diffractometry is subject to stringent requirements. Most X-ray tubes provide a linear source of X rays, approximately 1 cm in length and 0.1 cm, or less, in width. The area of the source may be further defined by operating techniques. If an isotopic source is to provide comparable operating conditions, the radioactive material must be laid down in an area of 0.1 to 0.2 cm^2. The area requirement immediately imposes a self-absorption limitation because of the relatively soft photons which are used in diffractometry (Mn K_α at 6 kev). The main difficulty with isotopic X-ray sources is that of providing useful photon intensity. Self-absorption may reduce the intensity below useful levels. Therefore the design of sources to minimize self-absorption is an extremely important part of this study.

The first source used was a 0.2- by 1.0-cm 100-mc ^{55}Fe source, which was prepared for us by the Isotopes Division of Oak Ridge National Laboratory. A description of the techniques by which this source was produced and a photograph of it are given in Ref. 1. The actual useful activity of the source, because of self-absorption, was about 13 mc. A diffractometer was constructed to accommodate this source and to allow for diffraction by powder samples. It was designed in the conventional diffractometer geometry, in which the source and detector lie on the circumference of a circle, with the powder sample at the center. The device is constructed so that the source remains stationary, while the detector rotates through angles of 2θ and the powder sample rotates through angles of θ. In this way the condition for Bragg diffraction is satisfied in accordance with the equation:

$$n\lambda = 2d \sin \theta$$

where n = order of diffraction
λ = wavelength
d = lattice spacing
θ = angle between incident radiation and a tangent to the surface of the powder sample

The diffractometer is illustrated in Fig. 1. Further illustrations appear in Ref. 2.

Initial calculations predicted a diffracted intensity of about 60 photons/min from the 200 plane of LiF powder. The signal-to-noise ratio for this low-intensity signal was improved by feeding the signal from the proportional counter, after suitable amplification, into a multichannel analyzer. In this way a signal from the 6-kev diffracted X rays would have to compete only with background radiation and noise that fell into the 6-kev energy region. Figure 2 shows a block diagram of the electronics.

A diffraction pattern (Fig. 3) was obtained with the system described above.[2-5] The intensities obtained were at about the predicted levels. Both NaCl and LiF samples were used initially; later, other crystalline materials were tested with satisfactory results. The powder was replaced with a single crystal of LiF, and a pattern (Fig. 4) was also obtained in this way.[6-8] These results were sufficient to establish that the technique of diffraction was possible with X rays from a radioactive source and were especially encouraging, since the ^{55}Fe source was relatively small. However, better results were needed in terms of both resolution and intensity. Improvement in resolution required a diffractometer more accurate than the one previously used, and an increase in intensity necessitated a more intense source with, hopefully, less self-absorption.

Camera Work

The incorporation of a radioactive source of X rays into an X-ray diffraction camera has been worked on independently by Lobov et al.[9-11] in Russia, and Semmler[12,13] and ourselves[14,15] in this country. Such a system takes full advantage of all of the benefits of a radioactive source. The unit is completely self-contained and portable, requiring no source of power. This is very attractive for applications such as geologic testing in remote areas. For use in the laboratory, the greater simplicity and minimum power, space, and cooling requirements ought not to be overlooked.

The first such related work appears to have been done by Lobov, who employed a reactor-produced ^{55}Fe source to obtain diffraction patterns from metal foils in a small, specially designed camera. He was hampered, however, by fogging of the film due to ^{59}Fe contamination inherent in his source. His device was a "Preston's Camera" based on back-reflection system rather than a Debye–Scherrer system.

Our own work centered upon the construction of a versatile diffraction camera with a 2-in. diameter. The source and film are mounted on the periphery and the diffraction sample is placed either on the periphery (in the focusing geometry position) or in the center (in the Debye–Scherrer geometry). Figures 5 and 6 show the details of the camera.

An attempt was made to obtain a back-reflection pattern by mounting a piece of iron foil on the camera periphery opposite to the source in the geometry of

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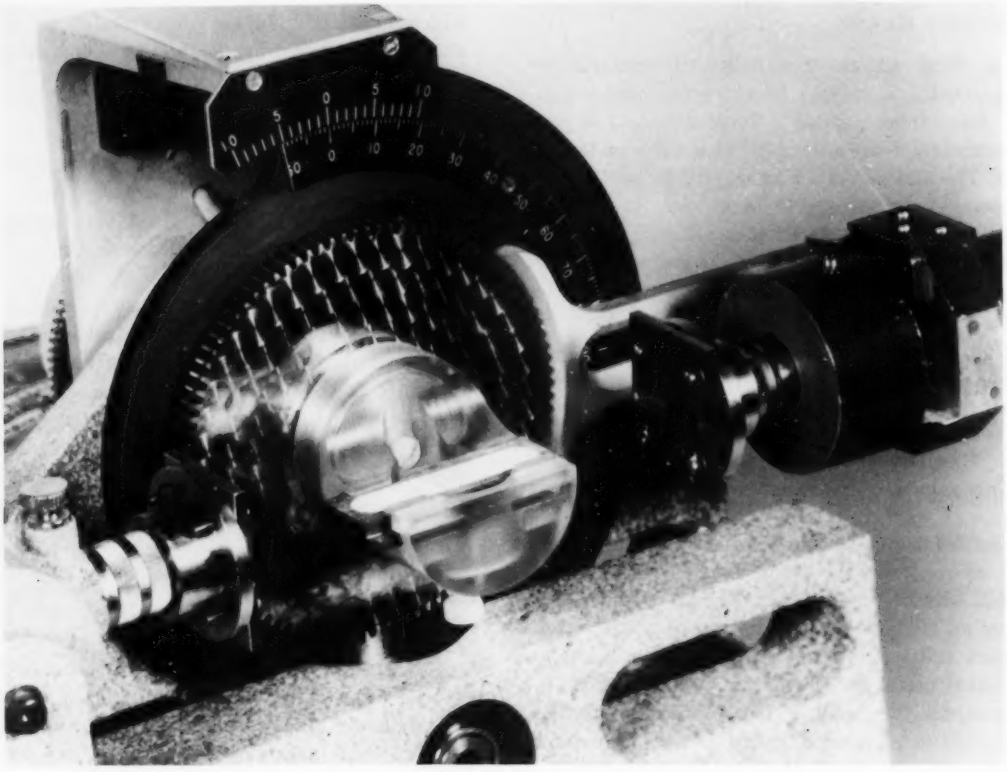

Fig. 1 The original diffractometer constructed on this project. The radioactive source is mounted in the housing shown at the extreme lower left. The diffracting sample is contained as a powder in the tray at the center. The detector assembly, which also contains the receiving slit, is at the right. The powder tray rotates through angles of θ, while the detector assembly rotates through angles of 2θ.

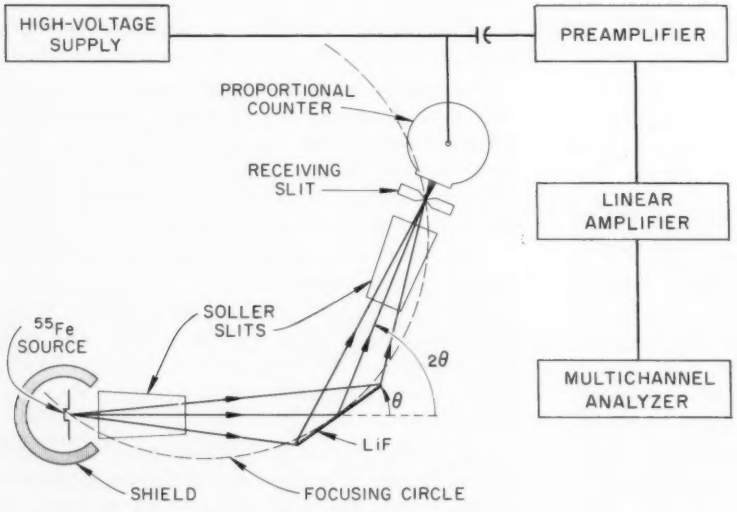

Fig. 2 The system and the electronics employed for the first powder diffraction done with X rays from a radioactive source. The effective source strength was about 13 mc.

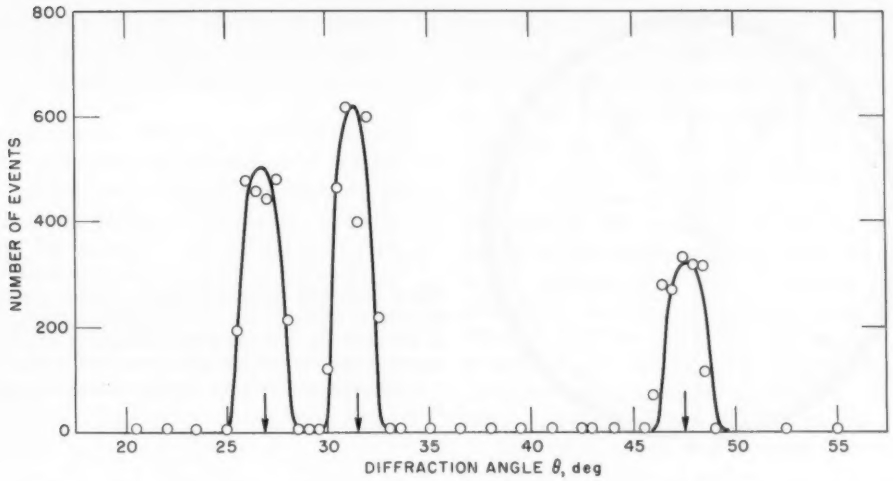

Fig. 3 The first powder diffraction pattern obtained with X rays from a radioactive source. The three peaks represent reflections from the 111, 200, and 220 planes at the angles 26.9, 31.5, and 47.6°, respectively. Each point represents counts accumulated in 50 min with the source in place, followed by a background subtract for 50 min with the source removed. The resolution for the peak from the 200 plane is about 2° FWHM intensity. The heavy arrows show the predicted diffracted position. [This figure first appeared in *Applied Physics Letters*, 9: 159 (1966).]

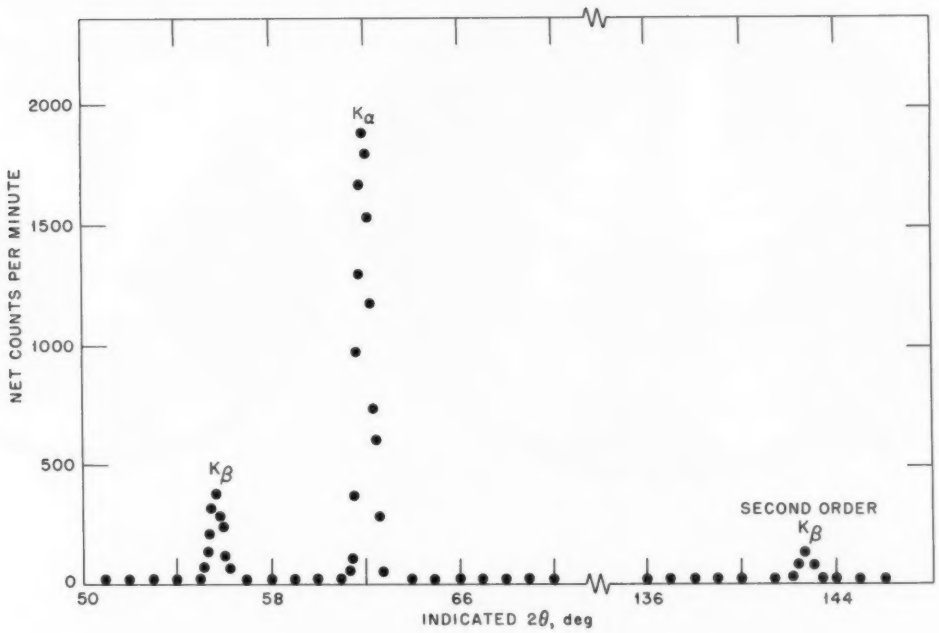

Fig. 4 A spectrum from the 200 plane of a LiF single crystal. The first order K_α and the first and second order K_β reflections are clearly visible. The resolution for the K_α peak is about 0.3° FWHM. The data were collected with a 256-channel analyzer by counting for 1 min at each angular position with the source in place and then doing a background subtract for 1 min with the source removed.

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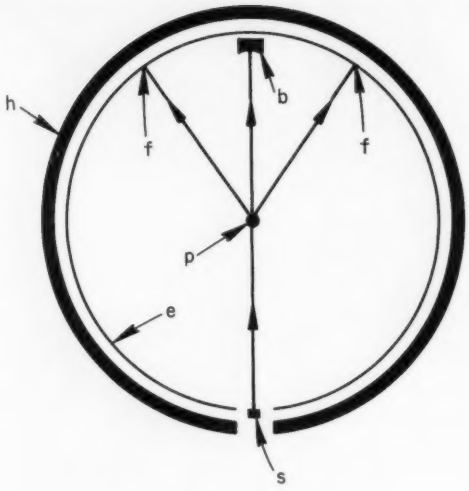

Fig. 5 The small diffraction camera that was constructed for use with a radioactive source; *s* is the source, *h* is the housing, *e* is the emulsion, *p* is the powder sample mounted in a glass capillary, and *b* is the beam stop; *pf* is the diffracted beam, and *f* is the point at which the diffracted beam is imaged on the film.

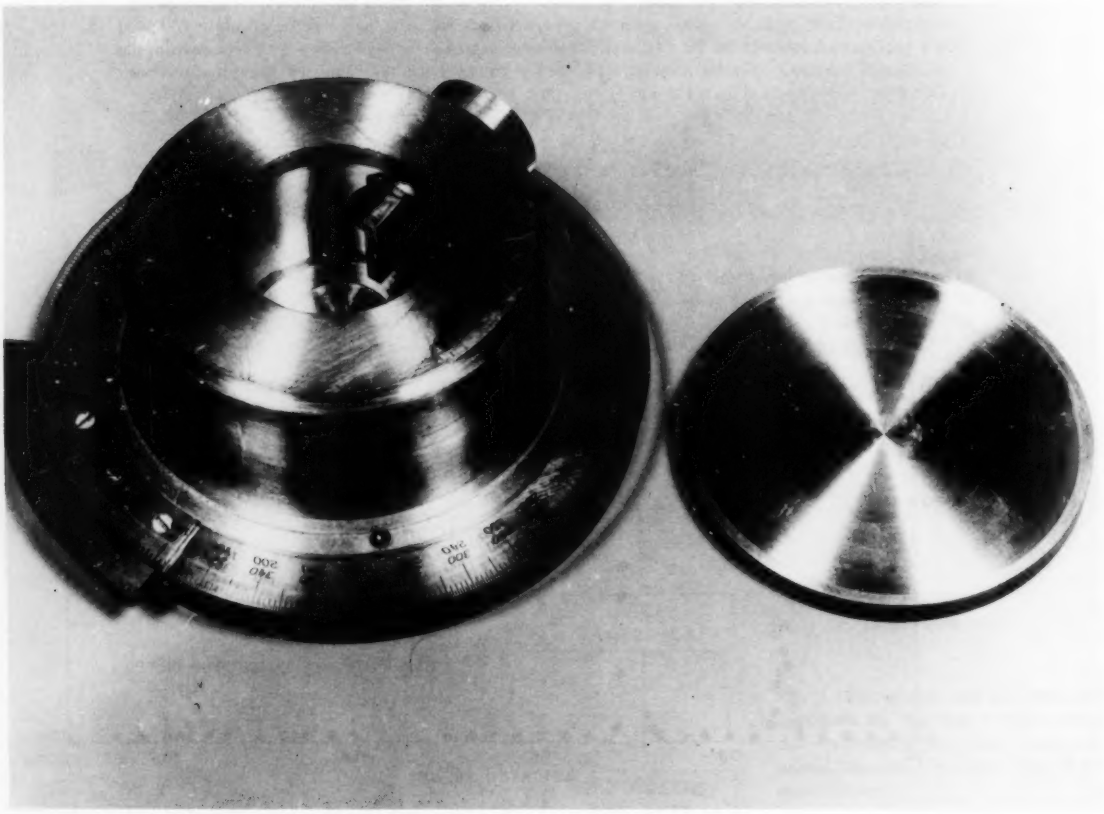

Fig. 6 The small diffraction camera. It is made of brass with a 2-in. inside diameter. The source housing and collimator can be seen protruding into the diffraction chamber from the top of the picture. The lightproof cover is shown at the right.

the focusing camera. Figure 7 shows the pattern obtained after 90 min. The lines (one on either side of the center) correspond to diffraction from the 211 plane of the foil.

We next attempted to obtain a powder diffraction pattern, since, if the device were to be used for analysis of unknown samples, this would be the technique of choice and the preferred form of the sample. A sample of LiF powder fine enough to pass a 200-mesh screen was placed in a thin-walled glass capillary and mounted in the center of the camera. After an exposure of 118 hr, the Debye–Scherrer diffraction pattern shown in Fig. 8 was obtained. This is, to our knowledge, the first analytical Debye–Scherrer powder diffraction pattern ever produced with X rays from an electron-capture isotopic source.

Current Investigations

Partially as a result of this successful preliminary work, Oak Ridge National Laboratory undertook the construction of a second ^{55}Fe source. The electrodeposited source was 0.2 by 1.0 cm with a total activity of about 200 mc. The self-absorption of this source turned out to be remarkably low, and the actual output of the source was about 15 times that of our previous source. With this more intense source, it was hoped to improve greatly the intensity of the diffraction patterns. We attempted, further, to improve the resolution and accuracy by replacing the laboratory model diffractometer with a Norelco wide-angle goniometer. A diffraction pattern from the LiF crystal, obtained with the new source and diffractometer, is shown in Fig. 9. The intensity was sufficient to allow use of very small divergence and receiving-slit openings and also to drive the detector continuously at a speed of 0.125°/min and record its output on a chart recorder.[16] A diffraction pattern from LiF powder obtained in the same manner appears in Fig. 10.

At the present time, modifications are being made to existing diffraction cameras to allow the use of the new source. Here, also, we expect to improve the reso-

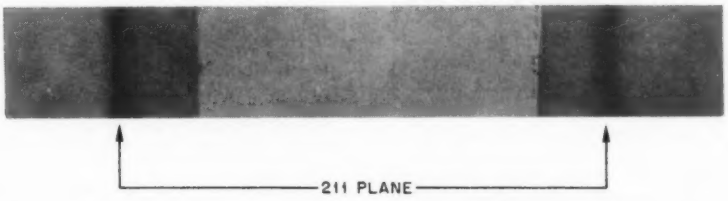

Fig. 7 A back-reflection diffraction pattern from the 211 plane of a piece of iron, obtained using ^{55}Fe X rays. The iron foil was mounted on the periphery of the diffraction circle, and the exposure was for 90 min. Exposures were generally short with metallic samples.

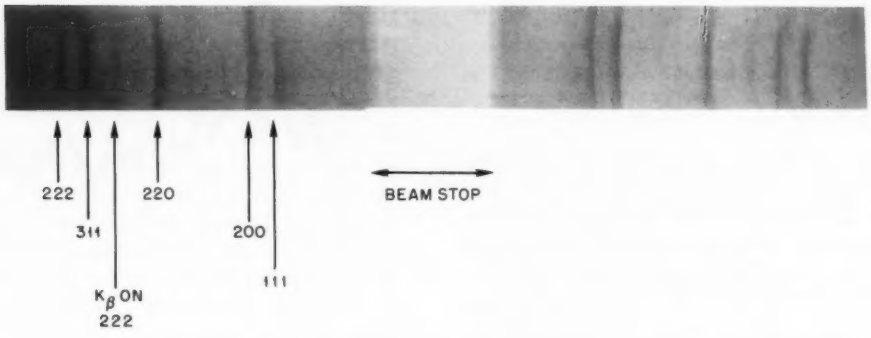

Fig. 8 Debye–Scherrer powder diffraction pattern of LiF powder taken in the camera using ^{55}Fe X rays (Mn K_α). The powder was mounted in a thin-wall glass capillary at the center of the camera. The exposure was for 118 hr. The lines represent reflections from the following planes (starting from the extreme left and reading toward the center): 222, 311, 220, 200, and 111.

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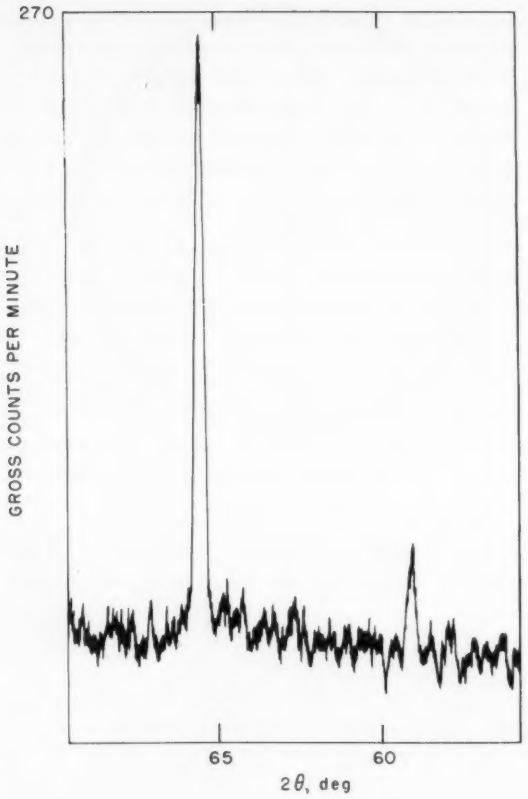

Fig. 9 An automatically recorded diffraction pattern of ^{55}Fe X rays on the 200 plane of a single crystal of LiF. The two peaks are due to the K_α and $K_{\beta 1}$ X rays. The scan speed was 0.125°/min and the chart speed was 3⅓ in./hr. The source was 200 mc. Angle 2θ is indicated, not absolute degrees.

lution (line-width separation on the film) and shorten greatly the exposure times.

Summary

The state of the art of the use of radioactive sources of X rays in diffractometry may be summarized by the statement that the X-ray intensities currently available from the ordinary beta-excited source are too low for feasible application. The diffraction phenomenon with such excited sources may be demonstrated, but low beam intensities call for extremely long exposures and relatively complex instrumentation to bring the pattern out of the background. Only a significant breakthrough in the construction of beta-excited X-ray sources, such as the achievement of beta emitters with ultrahigh specific activities, can make this X-ray source competitive with electron-capture sources such as ^{55}Fe. Improvement by a factor of 50 to 100 is needed. Electron-capture X-ray sources, on the other hand, hold considerable promise. Iron-55 is a prime candidate for such work and has been used successfully in our studies. Further investigation with ^{55}Fe beyond these preliminary studies is called for.

The advantages of the isotopic X-ray source, particularly ^{55}Fe, are considerable and include stability of output, monochromaticity (in the case of ^{55}Fe), a zero requirement for X-ray power, compactness, and lightness (a source is ordinarily about as large as a dime). A single disadvantage plagues all isotopic sources—the problem of intensity. In no case has an isotopic source been made that has the intensity of X rays equivalent to that of the ordinary X-ray machine (at the energies discussed herein). The use of ^{55}Fe makes possible the production of a source that has an intensity within a factor of 50 of the small X-ray units that are designed for portable use or remote-area application where power is limited or nonexistent. Much of this intensity problem hinges upon the question of resolution. Adequate intensity (with somewhat extended exposure times) can be obtained with the ^{55}Fe sources, provided that some loss in resolution can be tolerated.

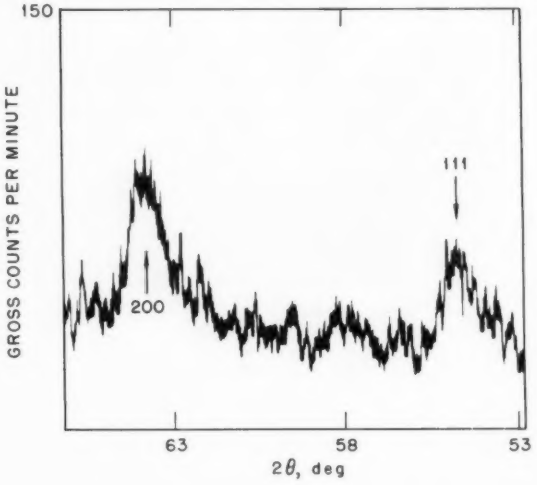

Fig. 10 An automatically recorded diffraction pattern of ^{55}Fe K_α X rays with a powder sample of LiF. The peaks represent diffraction of the K_α X ray in the 220 and 111 planes of the powder. The source was 200 mc. Angle 2θ is indicated, not absolute degrees.

We are of the opinion that source design is most important to success in this work and the effort devoted to it should be expanded. With improved source and diffractometer design, ^{55}Fe sources may provide competitive intensities, and acceptable resolution will still be obtainable with such a source. A source of 2 curies of ^{55}Fe with remarkably low self-absorption has already been produced. Such a source specifically designed for diffractometry work would improve intensity values almost to competitive levels. Promethium-147, on the other hand, when laid down in diffractometry geometry for an X-ray source, begins to show deleterious degrees of self-absorption. The beta-excited source, therefore, appears to be self-limiting because of this absorption problem.

Accelerator-produced ^{55}Fe has a distinct advantage over reactor-produced ^{55}Fe in that it may be made with a minimum of stable contaminating elements and radiocontaminants. The superiority of this ^{55}Fe, when laid down in the linear X-ray source geometry required for diffractometry, is unquestioned. The problem of cost of the ^{55}Fe source is a serious matter that is directly tied to accelerator time and appears to be increasing rather than decreasing. Mass production of ^{55}Fe by this technique could result in the highly desirable reduction of the millicurie cost. Certainly the ^{55}Fe source will not be used if 2 curies of the material costs as much, or more than, a good X-ray generator.

At the present time the beta-excited source for X-ray diffraction does not appear to be useful. The ^{55}Fe, however, even at this early date in the development, exhibits useful attributes for remote-area work and for usage in which electric power, space, and mass must be limited. Some of these applications could be in lunar exploration, in remote, "out-back" areas, and in physical locations where the highly fragile X-ray tubes cannot be used. Miniaturization is possible with the ^{55}Fe source, and the advantage of this should not be overlooked.

Finally, we stress again the interesting possibilities of the Debye–Scherrer camera for studies with an ^{55}Fe source. We have already demonstrated an encouraging increase in resolution and shortening of exposure times for certain camera applications. We have done this with a very small (in terms of millicuries) ^{55}Fe source; with a source more intense by a factor of 10, exposure times would be obviously only a matter of a few minutes to an hour. Consequently, reasonably good identifications could be made. A more intense source will also allow for improved line resolution. Our work has been done without image intensification or film sensitization, and with these a zero-power system may be developed that will use the Debye–Scherrer geometry for analytical work (possibly with Polaroid film). An isotope-powered camera would be highly suited to remote-area applications where analytical diffraction must be carried out. Possibilities that come to mind are those of geological exploration and either manned or unmanned planetary stations in which an automatic sampling device could provide diffraction samples for a zero-power diffraction camera and isotopic X-ray sources. (FJM)

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^{252}Cf: A New Isotopic Source for Neutron Radiography

By W. C. Reinig*

Abstract: *The properties of ^{252}Cf that make it suitable for use as a neutron source in neutron radiography are described.*

Nuclear reactors are the usual source of neutrons for radiography, primarily because of their intense neutron beams. Much of the development work in neutron radiography has been directed at problems peculiar to the nuclear industry, and the laboratories engaged in this work have had ready access to reactors. Although there are still many applications for neutron radiography within the nuclear industry, potential uses in other industrial and scientific fields are increasing rapidly. However, in some cases, it is impractical to bring the equipment to a reactor to be radiographed; in other cases, it adds to the cost and time to make the tests. Therefore, if neutron radiography is to find widespread use, intense and transportable neutron sources are required which can be used in plants, in laboratories, and in the field.

Californium-252, an intense neutron-emitting radioisotope, is a promising new source for radiography. A gram of ^{252}Cf will emit 2.34×10^{12} n/sec by spontaneous fission. The only other isotopic source with a high neutron yield which has attracted interest for neutron radiography[1,2] is ^{124}Sb–Be. The characteristics of ^{252}Cf and ^{124}Sb–Be sources with a yield of 5×10^{10} n/sec, which is adequate for radiography, are compared in Table 1.

Five thousand curies of ^{124}Sb is required for an emission rate of 5×10^{10} n/sec, while only 11 curies of ^{252}Cf is needed. The ^{124}Sb–Be source produces neutrons inefficiently by the action of the ^{124}Sb decay gamma on beryllium. A well-designed ^{124}Sb–Be source, such as described by Hennelly,[3] will emit 10^7 n/(sec)(curie); in contrast, ^{252}Cf emits 4.4×10^9 n/(sec)(curie). The very small volume of the ^{252}Cf source provides essentially a point source of neutrons. The comparatively large volume of the ^{124}Sb–Be source is due to the large amount of beryllium required for an efficient source. When good imaging techniques for fast neutrons are developed, a point source will provide geometric magnification to improve radiographic magnification, as is often done with X radiography.

Californium-252 neutrons have an average energy of 2.3 Mev; the neutron spectrum of ^{252}Cf is shown in Fig. 1. The 24-kev neutrons from ^{124}Sb–Be require less moderation for thermal-neutron radiography, but

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Table 1 Characteristics of ^{252}Cf and ^{124}Sb–Be Sources with Yields of 5×10^{10} n/sec

Source	Activity, curies	Volume, ml	Average neutron energy, Mev	Gamma dose rate at 1 m, r/hr	Half-life	Annual cost, $
^{252}Cf	11	<1	2.3	2.9	2.6 years	20,000
^{124}Sb–Be	5000	200	0.024	1.4×10^4	60 days	20,000

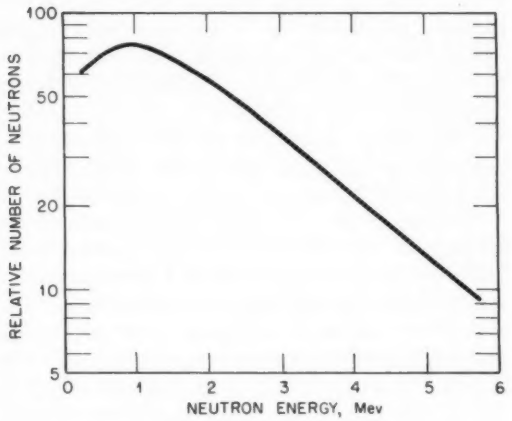

Fig. 1 Neutron spectrum of unshielded ^{252}Cf source.

the neutron energy of ^{124}Sb–Be may be too low for fast-neutron radiography.

The gamma dose rate from ^{124}Sb–Be is 5000 times as great as that from ^{252}Cf, and this high gamma radiation necessitates great attention to the protection of personnel during radiography. The magnitude of the gamma radiation is illustrated by this comparison: The dose rate a few inches from a typical 400-kvp industrial-radiography generator is 12 r/min, while that at 2 in. from a 5000-curie ^{124}Sb–Be source is 10^5 r/min. Some of the methods of detecting neutrons for radiography cannot be used in the presence of high levels of gamma radiation; other detection methods become complicated and expensive because of the need for gamma discrimination or reduction.

The annual costs of ^{252}Cf and ^{124}Sb to provide and maintain a minimum yield of 5×10^{10} n/sec are approximately the same, as shown in Table 1. These values, however, do not include fabrication and shipping charges. Because a californium source requires no target material, fabrication costs would be less than those for the ^{124}Sb–Be source. Shipping charges associated with replenishing the short-lived ^{124}Sb would be significant.

In addition to reactors and isotopic neutron sources, accelerators have been proposed as a source of neutrons for radiography. Sealed neutron generating tubes, developed in the past few years, with yields as high as 10^{11} n/sec, initially cost about the same as a ^{252}Cf source with the same neutron yield. Where a continuous source of neutrons is required, such as for radiographic inspection in industrial quality control, the cost per neutron from a ^{252}Cf source is considerably less than from an accelerator. The ^{252}Cf source requires no maintenance and can be transported easily to work areas or remote field locations. Also, the neutron emission from a ^{252}Cf source is constant over long periods of time, and it can be determined without supplementary measurements.

In summary, ^{252}Cf is a promising new neutron source for radiography. Although a few milligrams are now available to explore benefits of this radionuclide in radiography, several grams may be available in the early 1970's for developmental work. (MG)

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Use of Radioluminescent Materials in Space

By T. N. Lahr*

Abstract: *The uses of 3M Company's Microspheres—^{147}Pm radioluminescent devices—in the Apollo space program are described. These include a target for docking the lunar module and the command and service module, switch tips, a lunar landing indicator, and other markers. Some information is given on the background of the radioluminescent devices.*

Although low-level radioluminescent (R/L) materials have been used for many years, only recently have brightness levels been sufficient to make these materials useful for important projects. Existence of large quantities of inexpensive fission products has prompted companies to seek new uses for radioisotopes and has given impetus to development of greatly improved R/L materials.

One of the newer and more significant uses of R/L material can be found in the U. S. space program. Promethium-147, in the form of tiny inert beads called Microspheres, is serving as an activator for R/L material on the docking target of the Lunar Module (LM) designed to land this year on the moon. These Microspheres, mixed with phosphor, provide a long-lived reliable light to assist astronauts in docking and maneuvering in outer space. The docking target is made up of hundreds of these tiny beads which emit a soft, green light.

Properties of ^{147}Pm Microspheres

Promethium-147 is one of the most readily available fission products, being supplied by the U. S. Atomic Energy Commission in multicurie quantities at a relatively nominal cost. This isotope has a 2.7-year half-life and emits a beta particle with a maximum energy of 0.223 Mev and an average energy of approximately 0.07 Mev. These beta particles have a range of only about 0.025 in. in plastic, with shorter ranges in heavier materials. Because of this low energy and the ease of shielding of these beta particles, significant external radiation doses are not encountered with properly designed R/L devices using ^{147}Pm.

For any radioisotope, both the potential external hazard to a person near the source and the internal hazards to a person ingesting significant quantities of the radioisotope must be considered. The 3M Company has developed a process to convert the ^{147}Pm (and many other radioisotopes) into an almost totally insoluble form, 3M Brand Radiating Microspheres. These small ceramic particles can absorb and retain the isotope even in rather extreme environments. The particles themselves are completely inert physiologically and will pass through the body unchanged and be excreted almost 100%. The radioisotope sorbed by these particles will not behave in a normal metabolic way but will be retained by the Microspheres. In the past years, several animal studies have been completed which prove this point beyond a shadow of a doubt.

Because of the controllable particle size and easy handling of the Microspheres, they can be readily fabricated into products of almost any design. Product areas that are presently being pursued include radiation sources for industrial, aerospace, and medical use; static eliminators that use the alpha energy from ^{210}Po to ionize air and produce a conductive path to ground; and various R/L materials for use in the military, civilian, and aerospace fields.

Properties of R/L Materials for Spacecraft

Radioluminescent materials commonly consist of a physical mixture of a phosphor, which will luminesce when bombarded by electrons from an activator (usually a beta-emitting radioisotope), dispersed in a binder for application as a paint. Radioluminescent devices can also be prepared using certain radioisotopes in the gaseous form, but these sometimes have excessive bulk as well as problems of changes in state at the very low temperatures experienced in outer space. For aerospace applications, R/L materials that use ^{147}Pm in the form of 3M Brand Radiating Microspheres as the activator are of primary interest. Such materials offer several advantages: (1) They are reliable. No external power source is required, and there are no filaments that can break. The devices cannot be turned off except by complete destruction, and the components are functional at the temperature extremes encountered in space. There seems to be almost no way that the devices can fail. Qualification testing programs have been completed for various devices used on both the interior and exterior of the spacecraft. These devices were found to undergo every significant loss of function without any leakage of radioactive material.

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(2) The R/L devices require no power. As mentioned above, this leads to an increased reliability because of the absence of wires, filaments, etc. Further, this does not strain the already overtaxed power budget on most space vehicles. (3) Such devices are low in weight. Because of the lack of power-supply requirements, wires, etc., the use of R/L materials in no way increases the weight of the spacecraft. For example, the R/L switch tips, which are being provided to Grumman Aircraft Engineering Corporation (GAEC), weigh exactly the same as unlighted switch tips.

These three advantages of the use of R/L devices seem to recommend the free use of these items in spacecraft wherever they can do a suitable lighting job. However, the brightness of these devices is somewhat limited; that is, brightnesses in excess of 1 footlambert are not presently obtainable. This means that R/L devices are not suitable as floodlights or for very-long-distance use. The brightness levels which are required for the external docking maneuvers (presently about 0.2 footlambert) and for interior switch lighting (about 0.03 footlambert) are readily obtainable with presently available materials. Such devices definitely do have a shelf life, owing to both isotopic decay and possible radiation damage to the binder–phosphor system, but such shelf lives are in the 1- to 2-year range. This seems to be adequate for present spacecraft schedules and presently scheduled missions.

None of the above-mentioned limitations are intrinsic with R/L materials. These simply represent the present state-of-the-art products. Possibly higher brightness levels, as well as longer lives, may be achieved by using other activators and special radiation-resistant binder–phosphor systems.

Use of R/L Materials on Apollo Spacecraft

Various R/L materials are now being used on both the lunar module (LM) and command and service module (CSM) vehicles of the Apollo spacecraft. These R/L materials are used for both interior and exterior lighting, and in many cases are either the only lighting system used or a backup in the case of a darkside maneuver. Each of these uses will be described in turn. Uses for future missions will be suggested.

Exterior Uses

Lunar Module Docking Target. This device consists of a ring ($17\frac{1}{16}$ in. in diameter) and tee section containing radioluminescent disks (Fig. 1). Some 40 self-luminous "buttons" are placed along the black edging. Protruding from the center circle is a black tee bar ($10\frac{1}{4}$ in. long) lined with 26 more nuclear buttons. The radioluminescent disks, which contain approximately 300 mc of ^{147}Pm, have a brightness at time of

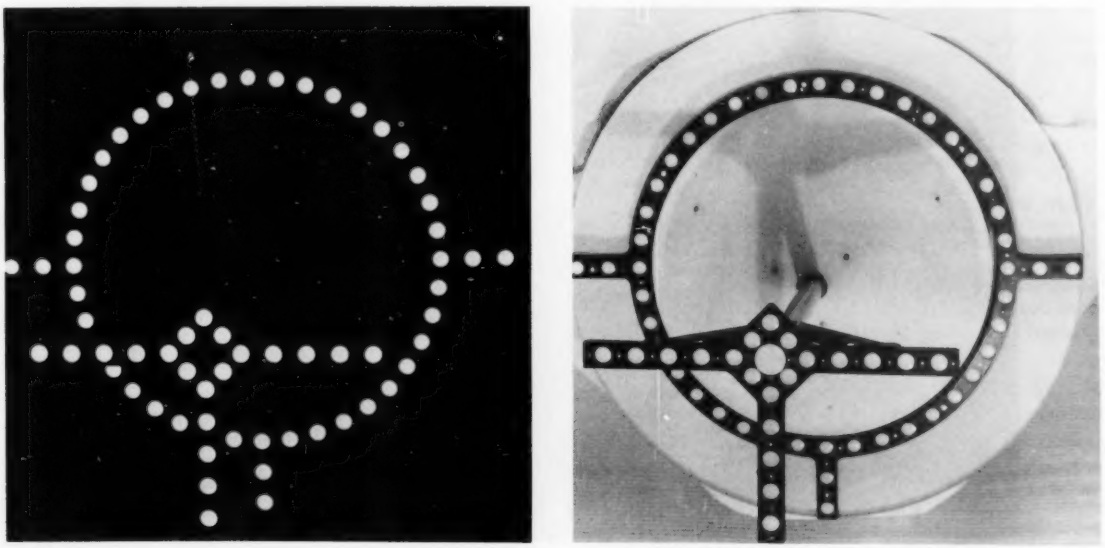

Fig. 1 Radioluminescent docking target used in Apollo space program. Target glows in the dark (left) and is painted in contrasting black and white (right) for daylight maneuvers.

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manufacture of approximately 1 footlambert. Under the −300°F (−184°C) conditions of outer space and after 1 year, the devices will have a brightness of approximately 0.2 footlambert. This is sufficient to allow easy visibility for completion of docking maneuvers at distances of 200 ft or more.

The docking target itself is constructed of aluminum sheet metal and is painted black and white to provide contrast for daylight visibility. When docking is carried out under standard conditions, the R/L disks form an image of the cross and tee, and docking can be accomplished without any additional lighting. Whether docking occurs in light or darkness, the astronauts will not see the actual hookup because of their position in the spacecraft. Without using the bull's-eye and optical sight, docking would be like trying to touch bumpers—or trailer hitches—while driving down a highway. The motorist can't see over the automobile hood, and the astronaut can't see over the front of his spacecraft.

This target is used principally during the first docking maneuver when the two spacecraft are coasting toward the moon. The command module pulls away from the LM that is contained in the adapter, turns around, comes back, docks with the LM, and extracts it from the adapter. During this critical docking maneuver, the pilot of the CSM lines up his vehicle using the reticle on his window (Fig. 2) in conjunction with the docking target that is mounted on the LM.

The 3M Company has not only developed and qualified the R/L disks but has worked with GAEC in the assembly and vibration testing of the flight hardware docking targets. Presently 3M is completely assembling these targets under contract to GAEC. Since this particular type of target is suitable for this

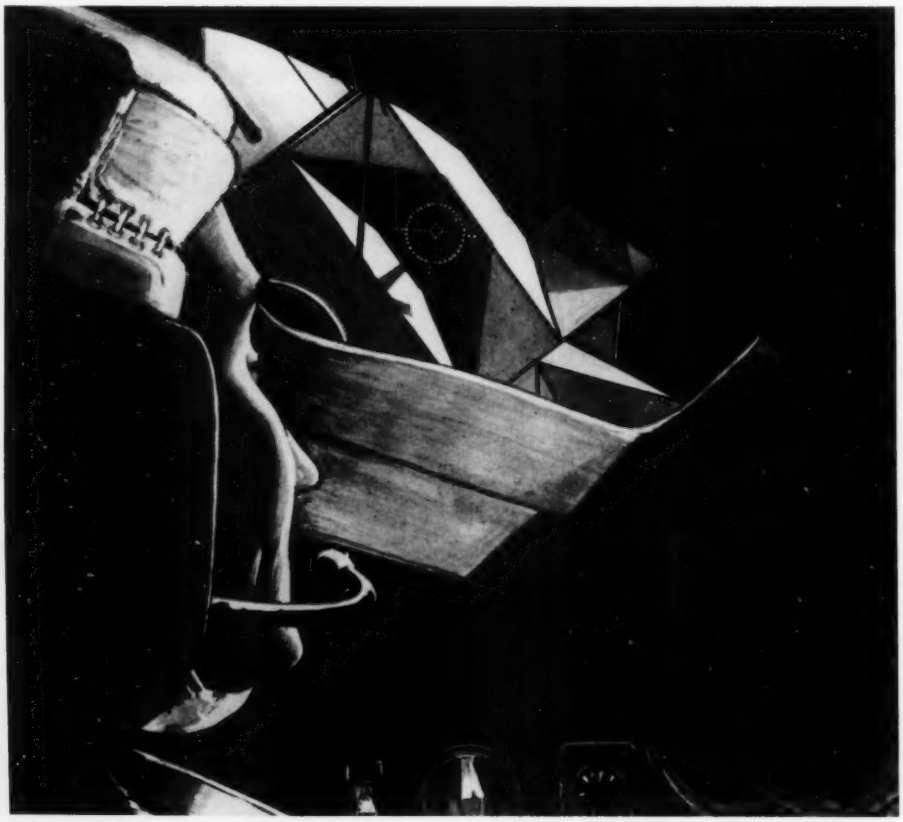

Fig. 2 Illustration of command and service module pilot's view of radioluminescent docking target on the lunar module.

docking maneuver and has been qualified for use in space environments, its use wherever a docking target is required would be reasonable.

Command Module Exterior Lighting. The command module vehicle will also have situated about its exterior several items that will be lighted by 3M R/L devices. These include the forward docking ring (around the hatch) that will be used for medium-range acquisition and docking maneuvers when the LM returns from the moon surface to the command module. Just as in the LM docking target, this ring will be built up using the R/L disks that have been qualified by 3M. Further, North American Aviation has designed several other items for use on the outside of the command and service modules. These include the marker for the step or handhold used by the astronauts for extravehicular activity as well as other markers, e.g., for hatch covers.

Interior Uses

Lunar Module Switch Tips. Because of the difficulty of supplying floodlighting to the inner portion of the LM, GAEC has gone over entirely to the use of R/L switch tips. These switch tips are fabricated by molding plastic caps to conventional toggle switches, drilling small holes therein, and inserting and sealing R/L material in the holes. The pattern of R/L material on various switches can show not only the the position of the switch and whether it is "on" or "off" but also its function. Four different designs are being fabricated. These include momentary and maintained toggle switches, a four-way switch, and a lever-lock switch. About 125 of these switch tips are used in each LM vehicle. These switch tips do not constitute a backup lighting system; they are the primary lighting mode for the switches on the interior of the LM. These switch tips have been put through a vigorous qualification testing program and have completely qualified in all interior LM environments.

Window Landing Scale. A landing-point designator also has been installed on the windows of the LM. Consisting of exact scales and degrees, it is designed to enable accurate moon landings. At about 5000 ft from the moon, the LM's commander will use the window scale to pinpoint the exact landing spot.

Conclusions

The properties of R/L materials, especially those activated with ^{147}Pm in the form of ceramic Microspheres, as well as their use in Apollo hardware, suggest that serious consideration be given to using R/L materials wherever a difficult low-level lighting problem arises. As an example of this type of problem, Douglas Aircraft is considering the possibility of lighting a tunnel in the Manned Orbiting Laboratory with these materials. Here, R/L disks would simply be mounted about the tunnel to guide the astronauts through it and to provide low-level lighting for the tunnel interior. The uses of this material can be many and varied and are limited only by the imagination of the designer. A significant design capability exists at 3M Nuclear Products owing to close association with both GAEC and North American. While the use of R/L devices is simple in concept, proper attention to design parameters is important. For this reason, 3M Nuclear Products should be consulted at the earliest possible point when these materials are being considered, so that they may extend the maximum possible help.

When the use of any radioactive material is proposed, an automatic reaction of fear or concern is often manifested. This is natural because of the usual context of "radioactive" as synonymous with "atomic bomb." Because of this natural concern, the use of ceramic Microspheres as the initial containment for the radioactive activator is of so much importance. Also because of this natural concern, the Manned Spacecraft Center at Houston, Tex., has contracted with Tracerlab/West for a safety analysis report for R/L sources used on Apollo spacecraft. This was submitted to the Manned Spacecraft Center in response to Task Order No. 18 of Contract NAS9-5117 on Jan. 20, 1967. This particular analysis considered all the R/L material on the LM and CSM spacecraft described above. Although this analysis is much too lengthy to recapitulate here, the conclusions reached are worthy of repetition. The most important conclusion was as follows:

> One general conclusion is of overriding importance; the use of ^{147}Pm microspheres in self-luminous discs cannot, under any creditable circumstances, cause exposure of biological significance.

This conclusion indicates that no special significance need be attached to the use of ^{147}Pm R/L devices in spacecraft. The functional usefulness of these devices should be the sole determining factor for decisions regarding their use. Unfounded concern about radiation and simple prejudice against the use of radioactive materials should have no place in such decisions.

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Current Trends in Labeled Compounds

By Seymour Rothchild*

Abstract: *The use of isotopically labeled compounds has grown continuously during the past 20 years. A large number of labeled chemicals have become available from commercial sources. Recent developments in labeled compounds have come about in tritium labeling, the biosynthetic method of preparation, labeled peptide synthesis, the application of chromatographic methods, and the problem of self-radiolysis.*

The use of labeled compounds for tracing dates back to Hevesy's work of almost 50 years ago. Gaining momentum in the late 1930's with the introduction of deuterium and cyclotron-produced isotopes, the large-scale usage of labeled chemicals developed when large amounts of relatively inexpensive reactor-produced radioisotopes became available in 1946. Commercial sources for labeled compounds, which were tagged primarily with ^{14}C, multiplied rapidly in the ensuing years. However, the major impetus to the usage of labeled compounds developed in the mid-1950's because of the mushrooming of grants to biomedical investigators. The commercial labeled-compound industry in the United States has grown to the present $9 to $10 million annual sales rate with six major suppliers and several smaller ones. Carbon-14- and ^{3}H (tritium)-labeled compounds account for well over 90% of the sales volume, not including the application of radiopharmaceuticals to clinical diagnosis.

The price for barium carbonate-^{14}C, which is the raw material for further synthesis of carbon-labeled compounds, was $50/mc in 1948 and has tumbled to less than $6/mc today. During the same period, in which the volume of production increased by factors of 10, prices for labeled compounds have fallen to one-half to one-tenth of the original prices. A significant change in the market took place about 1956 when the Oak Ridge price of tritium gas dropped from $100 to $2/curie. Coincidentally, liquid scintillation counters, which greatly simplified the measurement of tritium, began to appear in significant numbers. Also, in 1957, Wilzbach announced the results of his tritium-gas-exposure method for labeling.[1] These developments accelerated the use of tritium in tracer work.

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The present commercial dollar sales volume of tritium-labeled compounds is estimated to be about one-third that of ^{14}C-labeled compounds. Investigators have found several important advantages in the use of tritium as a tag: higher specific activity, lower cost, and better resolution in autoradiography.

The use of labeled compounds during the past decade has been affected by improved methods of synthesis and a growing awareness of the factors affecting the purity of labeled compounds. As the applications of labeled compounds become more sophisticated, investigators are increasingly concerned about the effects of impurities on the validity of their work. Some of the more significant recent developments in the field of labeled compounds are outlined below.

Tritium-Labeling Techniques

The use of a suitable precursor compound for reduction remains a prime method for obtaining a tritium-labeled product, especially at high specific activity (> 10 curies/millimole), except for some aromatic substances that are readily labeled by catalytic exchange. The reducing agents employed generally are tritium gas (58 curies/millimole), sodium borohydride-^{3}H (up to 6 curies/milliatom of hydrogen), or lithium aluminum hydride-^{3}H (up to 50 mc/milliatom of hydrogen). Precursors include olefins, acetylenes, organic halides, nitriles, and carbonyl compounds. Nonspecific labeling patterns have been observed[2-5] in heterogeneously catalyzed tritium-gas reductions, and the resulting products, therefore, are best described as nominally labeled at the site or sites of reduction, unless otherwise demonstrated to be specifically labeled. The more critical view of labeling sites, however, is not surprising in view of the massive amount of isotopic exchange studies[5] performed in the last 30 years. The stereochemistry of reductive dehalogenation,[4] desulfurization,[6] and dehydroxylation[7-9] has received attention. A critical review[10] tying together heterogeneous catalytic reduction and catalytic exchange on a mechanistic level presents current theory and represents a high level of sophisticated thought.[10,11] The development of homogeneous catalysts for reducing systems promises other advanced tools of labeling.[12-14] The labeling

patterns and stereochemistry of labeled steroids resulting from the latter technique have been studied,[15] and data should be available soon.

Catalytic exchange techniques have advanced greatly in recent years. This type of tritium labeling, performed directly on the substrate of interest, offers great versatility since no precursor is required. Specific activities in excess of 1 curie/millimole are achievable; racemization of asymmetric centers and the extent of decomposition occurring during labeling are dependent upon the conditions used.

Catalysts employed include acids and acidic compounds (acetic acid, trifluoroacetic acid, boron trifluoride etherate, sulfuric acid, phosphoric acid, and alumina), bases, heterogeneous metal catalysts[16] on a variety of supporting materials, and homogeneous catalysts.[17] New methods of exchange using supported heterogeneous metal catalysts have been developed[18] for the labeling of materials too sensitive to radiation damage to be labeled under the more rigorous conditions required for tritium-gas exposure (prolonged exposure to radiation) and acid-catalyzed exchange (high temperatures). Thus, L-amino acids are produced at specific activities of >1 curie/millimole, some with less than 2.5% D-isomer.[18] The use of D-amino acid oxidase[19] on the product of the above exchange can, in some instances, produce an L-amino acid in a high state of optical purity.

The catalytic exchange of nucleosides and nucleotides has also been studied,[18] and these substances can be produced at specific activities from 1 to 15 curies/millimole. The sensitive triphosphate bond of the purine nucleotides survives the labeling technique to produce crude tritium substrates of exceptionally high quality. Other substances that are amenable to this catalytic exchange technique are sugars, polysaccharides, phenols, some catecholamines, steroids and related substances, and substituted amines.

Many substances that are insoluble in common solvents or are unstable in solution are still best labeled by the tritium-gas-exposure method of Wilzbach.[1,20] A mechanism has been advanced that explains the exchange process,[21] and many variations of the original technique have been developed.[20] One very useful variation is the silent discharge method[20] that has been used effectively to label the polypeptide oxytocin.[22] The products resulting from this type of labeling frequently are of lower specific activity (<100 mc/millimole) and require extensive purification. On the other hand, at this writing, this technique is still the simplest way to label biologically active polypeptides.

Biosynthesis

The increasing sophistication of tracer applications has led to the urgent need for more-complex labeled compounds, which are difficult for the organic chemist to synthesize. Fortunately biosynthetic methods have made many of these compounds much more accessible, and increasing attention is being focused on this approach to the preparation of labeled compounds.

Biosynthesis—in which enzymes, microorganisms, plants, and animals may be used—is ideally suited to the production of labeled compounds of high specific activity and optical purity. Though usually conducted on a semimicroscale or microscale, biosynthesis has the advantages of high yields, efficient control of by-products, stereospecificity, and the ability to produce chemically unstable biological compounds by virtue of the mild conditions used. While an exhaustive survey of the production of labeled compounds by biosynthesis is beyond the scope of this review, a brief listing will illustrate the range of these methods and indicate the possibilities.

A widely used method is the cultivation of unicellular organisms, such as green algae (*Chlorella pyrenoidosa* or *Euglena gracilis*) in a $^{14}CO_2$ atmosphere for the production of most of the ^{14}C uniformly labeled (U) amino acids,[23,24] fatty acids,[25,26] nucleosides and nucleotides from nucleic acids,[18,27] and a variety of carbohydrates.

Intact animals such as the rat have been widely used by introducing phosphate-^{32}P intravenously or intraperitoneally for the preparation of phosphorylated intermediates such as sugar phosphates, nucleotides, phospholipids, and coenzymes.[28] Similarly, the injection of glucosamine-^{14}C into the rat permits the isolation from the liver of uridine diphosphate (pyro)-*N*-acetylglucosamine-^{14}C (Ref. 29).

The cultivation of microorganisms has found wide application in the preparation of radioactive compounds. Particularly useful are genetically blocked mutants that require specific supplements for growth. For example, *Escherichia coli*$_{WC}$-, grown on uracil-2-^{14}C, can be used to form labeled uridylic and cytidylic acids in the ribonucleic acid (RNA), and thymidylic and deoxycytidylic acids in the deoxyribonucleic acid (DNA).[30]

Enzymes for the catalysis of in vitro reactions have been extensively employed. For example, chemically synthesized DL-mixtures of amino acids[31,32] are resolved with acylase, and radioactive sugars are converted with sugar kinases, utilizing adenosine triphosphate (ATP), to their corresponding sugar phos-

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phates.[33-35] The latter, in turn, can be enzymatically condensed with nucleoside triphosphates to yield a variety of nucleotide sugars.[36,37] Estrogens can be made from neutral steroids by use of aromatizing enzymes that have been isolated from the placenta,[38] and tetrahydrocorticoids are conveniently made by use of enzymes isolated from rat liver.[39] Rat ovary is the source of reductase enzymes employed for the production of 20 α-hydroxysteroids, while sheep blood,[40] which contains isomerase, is used for the preparation of epitestosterone.

Intact plants, e.g., *Canna indica*, have been used to produce carbohydrates-^{14}C such as sucrose-^{14}C (U) (Ref. 41) from $^{14}CO_2$, while red seaweed (*Iridothycus flaccidum*) produces high yields of galactose-^{14}C (U) (Ref. 42). Tobacco leaves produce starch-^{14}C (U) (Ref. 43) and sterols such as stigmasterol-^{14}C and sitosterol-^{14}C (Ref. 44), and feeding foxglove leaves with pregnenolone-7-^{3}H has provided labeled glycosides, such as digoxin, digitoxin, and gitoxin.[45]

Peptide Synthesis

The conventional process for the chemical synthesis of biologically active polypeptides is a slow, exacting procedure. It involves the coupling of suitably blocked amino acids to form small peptides that are isolated and purified at each stage. This process is repeated over and over again until the polypeptide is obtained, usually in extremely small yields. The difficulties encountered with the usual methods of synthesis, which are compounded manyfold by the introduction of labeled reagents, have severely limited the availability of isotopically labeled polypeptides, with the exception of those generally labeled with ^{131}I or ^{3}H.

Classical methods of peptide synthesis have been used for the incorporation of leucine-^{3}H into oxytocin,[46] while tyrosine-3-^{3}H has been incorporated into both oxytocin and lysine-vasopressin.[47] Reductive dehalogenation of the iodinated tyrosine groups with tritium gas has been successfully used to label oxytocin,[48] and the spark-discharge method[49] and the Wilzbach gas exposure technique[1] have been used for the preparation of tritium-labeled angiotensin II (Ref. 22). Both of the latter techniques yield relatively high-specific-activity products, but their usefulness is limited because of the formation of high-specific-activity impurities that require elaborate and often difficult procedures for their removal. The difficulties associated with the synthesis of specifically labeled polypeptides by conventional means as well as those associated with the exposure techniques have been successfully overcome by the use of the Merrifield solid-phase method of peptide synthesis.[50]

The Merrifield procedure operates by the ingenious technique of coupling the initial amino acid to a solid polymeric support that is insoluble in the solvents employed. The amino acid polymer, purified in excellent yield by simple filtration, is then coupled with the next amino acid. The filtration and coupling of each successive amino acid proceed alternately until the proper chain length and amino acid sequence have been reached. The desired soluble peptide is then removed from the solid support and purified by conventional methods. The advantages of the procedure are the reduction of the number of multistep processes, the coupling of amino acids without side reactions, the use of specially adapted blocking agents, high yields, and a relatively low percentage of racemization. These advantages make the solid-phase method ideal for the preparation of radioactive peptides.

The method has been utilized for the preparation of [2,3-L-proline-^{14}C (U)] bradykinin, which was found to be identical both chemically and biologically to known standards.[18] Other peptides made by this method are [2-L-proline-3,4-^{3}H nominally labeled (N)] bradykinin, [3-glycyl-2-^{3}H] glutathione, [1-L-aspartic-5-L-isoleucine-^{14}C (U)] angiotensin II, and [1-L-aspartic-5-L-isoleucine-10-L-leucine-4,5-^{3}H (N)] angiotensin I (Ref. 18). More recently the synthesis of [5-L-isoleucine-10-L-leucine-^{14}C (U)] angiotensin I has been reported.[51]

This modest list is but a portent of future accomplishments in the field of labeled polypeptides. This method may conceivably be the key that will make possible the preparation of complex radioactive polypeptides, such as growth hormone, insulin,[52] and ferredoxin.[53]

Chromatography as an Analytical Tool

Chromatography has shown the most dynamic development of all the techniques applied to the analysis of labeled chemicals. Generally, chromatographic procedures are well suited to and, indeed, are very efficient for the analysis of the microquantities that can be readily detected in the radioactive form. The detection of radioactivity on chromatographic media, e.g., paper or thin-layer, has historically been accomplished by scanning, removal or elution for counting, or by autoradiographic procedures. All these methods have been updated to some degree in recent years. Although scanning devices for paper chromato-

grams or electrophoretograms have remained essentially unchanged, modifications that adapt these instruments to the scanning of thin-layer plates have become popular. Attesting to the wide acceptability of thin-layer chromatography (TLC) has also been the recent appearance of at least one unit marketed principally for the scanning of plates[a] and adaptable for two-dimensional thin-layer chromatograms. A unit with multiple detectors designed for a rapid "picture" of the distribution of activity on a plate has recently been announced.[b] Several publications on the evaluation of scanning in general and of plates in particular have recently appeared,[54,55] and numerous scanners have been built and described in the literature.

The detection of radioactivity on thin-layer plates by all currently used methods has been the subject of a detailed review by Snyder.[56] Zonal scraping followed by liquid scintillation analysis was found to be an excellent procedure capable of high resolution and sensitivity.[57] The commercial availability of a semiautomated device for the accurate removal of layers[c] should make this an extremely attractive method, especially in laboratories with liquid scintillation equipment and where the ultimate in sensitivity is required.

Autoradiographic procedures have been dramatically extended to tritium by the application of scintillation principles. The conversion of radioactivity into light has been accomplished by placing a fluor close to the chromatographic media by spraying from solution,[58] by incorporation into the adsorbent during plate preparation,[59] or by exposure of the film while it is immersed in a solution of the scintillator.[60] A very recent development has been the marketing of a scintillator formulation combining the spray convenience with the durability and efficiency of a plastic film.[d] The high sensitivity attainable with these autofluorographic systems is due, in large part, to the observation of Luthi and Waser[59] that a dramatic increase in fluor efficiency can be obtained as the temperature is lowered. Autofluorography, combined with the careful application of standards to minimize the problems of film nonlinearity, promises to be very useful for the evaluation of purity of tritium-labeled compounds.

The application of activity monitoring systems to gas–liquid chromatography (GLC) has recently come into popularity. The last few years have seen a significant increase in the utilization of continuous flow systems that incorporate either proportional counting tubes[e,f,g] or ionization chambers[h] for detection. In addition, efficient procedures and equipment[i] are now available for trapping volumes that contain peak amounts of activity and assaying by liquid scintillation counting. Karmen[61] has comprehensively reviewed and compared the various procedures available for GLC. The application of GLC techniques has resulted in a definite upgrading in the purity of volatile radiochemicals as well as some new insights into the evaluation of high-molecular-weight compounds that can be readily converted into volatile derivatives, e.g., steroids, carbohydrates, and fatty acids.

The phenomenon of isotopic fractionation has been observed with such procedures as paper, column, gas–liquid, and thin-layer chromatography. Klein[62] has reviewed the field, and papers have appeared showing the degree of separation encountered in everyday analysis. The importance of these observations, particularly in double-label experiments, is apparent, and greater practical use of the effect is expected in the next few years.

There has been an increasing recognition that chromatographic "artifacts," i.e., breakdown of the labeled compound during analysis and adsorption losses, may occur during the purity evaluation.[63] Although both these phenomenons have always been a problem in the general analysis of trace compounds (notably, pesticides and catecholamines), they are of particular importance today owing to the increasing demand for higher and higher specific activity radiochemicals. Of additional concern are the practical economics of the not insignificant losses that may occur when an analytical system is adapted to preparative purposes. Methods for overcoming these problems that have been discussed include pretreatment of the media to remove materials that catalyze the breakdown,[64] the use of multiple systems to detect and

[a] Berthold Radioactivity Scanning System, Varian Aerograph, Walnut Creek, Calif.

[b] Beta Camera, Baird-Atomic, Inc., Cambridge, Mass.

[c] Snyder-Kimble Automatic TLC Zonal Scraper, Analabs, Inc., Hamden, Conn.

[d] Omnispray, New England Nuclear Corp., Boston, Mass. (Omnispray is a registered trademark.)

[e] Radioactivity Monitoring System, Barber-Colman Company, Rockford, Ill.

[f] Biospan Gas Radiochromatography Counting System, Nuclear-Chicago Corporation, Des Plaines, Ill.

[g] Gas Flow Proportional Detector System, Packard Instrument Co., Inc., Downers Grove, Ill.

[h] Radioactivity Analysis System, Varian Aerograph, Walnut Creek, Calif.

[i] Fraction Collector, Packard Instrument Co., Inc., Downers Grove, Ill.

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quantitate the breakdown,[65] and at least one study where an antioxidant was incorporated into the chromatographic medium.[66] There have been many other cases cited throughout the literature, and these effects must definitely be considered carefully in evaluating a method.

Self-Radiolysis of Radiochemicals

Prior to 1953 (Ref. 67), the low average energy of the beta emission of ^{14}C and ^{3}H was believed to cause little molecular damage to a radiochemical. As a result, the idea was accepted that there would be little practical difference between the stability of a labeled compound and that of its nonlabeled counterpart. The dynamic increase in the synthesis and use of high-specific-activity radiochemicals following this period and the consequent close examination of these compounds showed that they were indeed subject to marked changes because of self-radiolysis.[68,69] These findings prompted the appearance of a number of publications dealing with the theoretical aspects of radiodecomposition[68,70] as well as with the more practical aspects of preventing and decreasing this decomposition.[70,71] From these observations during the late 1950's and early 1960's, basic rules were developed for the storage of radiochemicals. At various intervals during this period, papers were published which listed long tables of radiochemicals that had been stored at various temperatures and usually arbitrary time periods.[71-74] These tables made it possible for the user of radiochemicals to ascertain what he might expect when storing his own radiochemicals under similar circumstances.

However, the decomposition rates reported by various investigators could almost never be duplicated. Often a researcher could not even duplicate the decomposition rates for different preparations of the same compound. Even worse, samples of the same lot of a radiochemical often displayed different stability or decomposition rates when stored by different investigators under slightly different conditions. In seeking answers to the problem of variability, a number of laboratories have recently reexamined existing practices for coping with radiodecomposition.[72-76]

The factor most responsible for the variability in decomposition rates is clearly that of chemical purity.[72,75,76] The accumulation of chemical impurities from synthesis and from solvents has been shown to have a considerable effect on radiochemical stability. Heavy-metal ions, such as iron and copper,[76] normally present in trace quantities in the nondistillable fraction of commercial-grade solvents, contribute to this decomposition. The variability in decomposition is now believed to be a result of the use of different grades of solvents during synthesis and storage. The incomplete removal of chemical impurities during purification also can have a major effect. The use of different chromatographic solvent systems or completely different purification methods such as distillation, crystallization, or column or paper chromatography can result in decomposition rates that differ remarkably for the same labeled compound.

The great emphasis on the removal of both the chemical (i.e., the nondistillable tars contained in solvents) as well as the radiochemical impurities arising from the synthesis has greatly extended the shelf life of many labeled compounds. These findings are negating the commonly accepted idea that high-specific-activity tritium-labeled compounds are much more difficult to store in solution than the ^{14}C-labeled compounds. These materials, when purified as indicated above, exhibited decomposition rates similar to those observed for their ^{14}C-labeled counterparts.

While low temperatures generally slow up the radiolytic decomposition rates, an increase in instability can result when these temperatures cause the storage solvent to crystallize.[76] The crystallization process may cause zonal localization of the solute in the solvent, thus effectively removing the beneficial protective matrix of solvent molecules. Current practice dictates that a radiochemical in an organic solvent be stored just above the freezing point of the solution. The proper ratio of binary solvents can give optimum conditions for the storage of labeled compounds.[76,77] However, the optimum conditions for each system must be determined separately.

Of interest is the observation that the decomposition rate of a radiochemical is not linear throughout its entire storage life.[76] Radiochemicals appear to exhibit a linear decomposition rate initially but proceed to decompose in an almost exponential fashion when the concentration of decomposition products reaches a critical point. The practical implication of these observations is that the investigator should regularly examine his radiochemicals during storage and repurify—for maximum protection—when the decomposition products reach a modest level (5 to 10%).

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New Radiometric Viscometer*

A viscometer that uses only one detector to measure the rate of sinking of a radioactive steel ball in a liquid has been described by Romanian scientists. The system is designed for use with very viscous liquids, e.g., molten glass or slag, paints, pastes, or liquid plastics.

The basic components of the viscometer (Fig. 1) are a radioactive source, which is 0.63 mc of ^{60}Co in the center of an 8-mm-diameter steel ball, and a Geiger–Mueller (G–M) counter. A lead filter is included to make it possible to adjust the count rate.

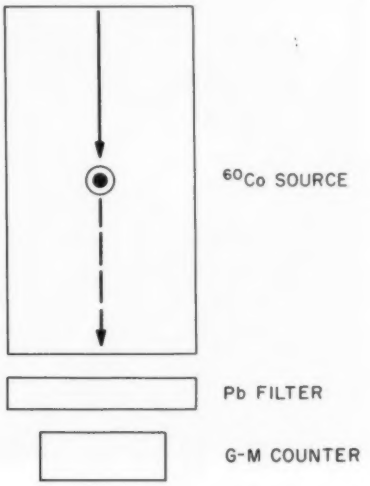

Fig. 1 Radiometric viscometer using a ^{60}Co source and a single G–M counter.

The liquid under study is contained in a vessel of 10 cm effective height. The source is allowed to sink in the liquid, and, as it drops, the count rate shown by the G–M counter is recorded. The rate of sinking of the source in the liquid can be determined using a previously established calibration curve relating count rate to vertical location in the liquid. In the experimental setup the counter was located below the vessel containing the liquid, but in an industrial plant it could be placed elsewhere if necessary.

The system geometry and electronics are simple, and the source activity is low enough that special shielding is not required. In tests at 25°C, the viscosity of silicone oil was determined with a precision of better than 2%. (MG)

*Abstract of "A New Improved Radioactive Viscosimeter," by H. N. Fuia and I. I. Ponner, *Buletinul Institutului Politehnic Gheorghe Gheorghiu-Dej*, **29**(1): 73-78 (1967).

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Radioisotopic Control for Pipeline Crawler

A pipeline crawler designed for X-radiographic inspection of circumferential welds in pipelines is controlled in its movement by a 1-mc ^{60}Co source. The source is positioned outside the pipe (Fig. 1) at the weld to be X rayed, and radiation detectors inside the pipe cause the battery-moved crawler to stop when the weld is reached. The film is automatically exposed to the 200-kv, 5-ma X-ray unit in the crawler, and the unit then moves on to the next weld. The crawler can examine a mile of pipeline before its Ni–Cd batteries have to be recharged. It is distributed by Picker Industrial of White Plains, N. Y.

(MG)

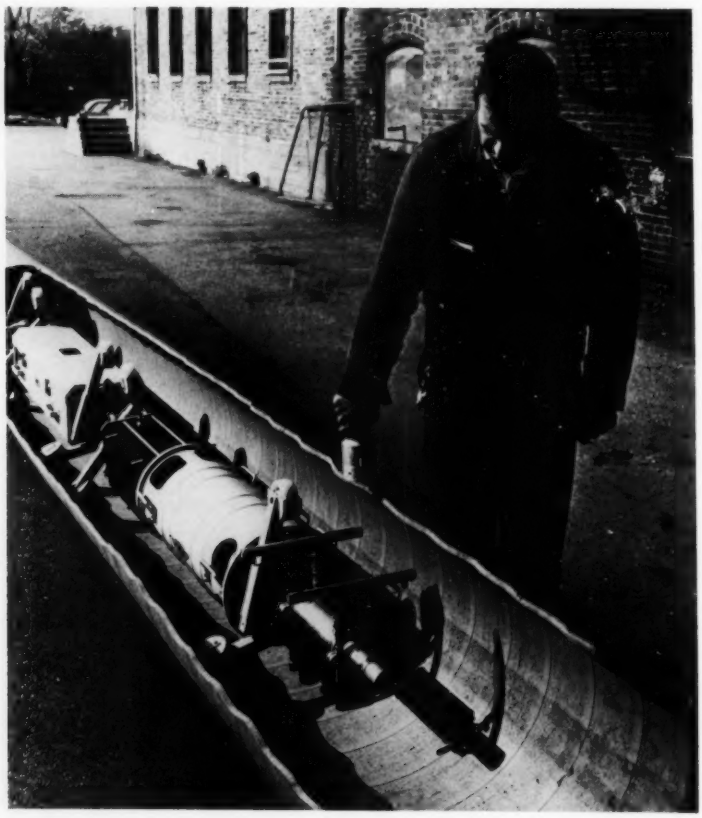

Fig. 1 Radioisotopically controlled pipeline crawler.

^{252}*Cf: Its Use and Market Potential*

A brochure on ^{252}Cf has been issued by the Savannah River Operations Office of the USAEC. With approximately 25 pages of text, numerous illustrations, and 45 references, this booklet summarizes the characteristics of ^{252}Cf and presents the AEC's ^{252}Cf market evaluation. The booklet may be obtained without cost by qualified scientists working in fields where ^{252}Cf is applicable. Requests for copies should be sent to the Manager, Savannah River Operations Office, P. O. Box A, Aiken, S. C. 29801. Supplements to the booklet will be issued periodically to review progress in the development of applications for ^{252}Cf. (MG)

Section

III

Isotopes and Radiation Technology

Food Irradiation

Food Irradiation as a Threat to Deep-Freezing Processes*

By J. Masefield†

Abstract: *The status of food irradiation as a process commercially competitive with deep freezing is examined. The use of radiation to depress growth, destroy insects, pasteurize, and sterilize is explained. The problems of obtaining approval from health authorities and of controlling organoleptic changes are pointed out. Food irradiation does not at this time present a threat to deep freezing.*

The preservation of food by treatment with ionizing radiation may emerge in the future as an important adjunct to existing food-preservation techniques. With a few exceptions, health authorities have been reluctant to approve food irradiation, and the process is not being used commercially. This discussion is limited to future feasibility and to the highly industrialized nations, although food irradiation may play its most important role in the developing nations where lack of electricity makes refrigeration of food impossible. In Western society, food irradiation will mean that the housewife can store refrigerated foods two to three times longer than at present and that prepackaged partly cooked sterilized items which are only available fresh now will be available off the shelf. Despite the potential threat of this new process, competition among freezing, pickling, canning, curing, and dehydration is likely to be more significant for many years than any economic threat from food irradiation.

*Summarized from a paper presented at an International Study Conference on Problems of Deep Freezing, Freeze-Drying, and Irradiation, November 6–8, 1968, at the Gottlieb Duttweiler Institute for Economic and Social Studies, Ruschlikon-Zurich, Switzerland.

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Interaction of Radiation and Matter

The underlying mechanism of food preservation by irradiation is that the energy of the photons or electrons is dissipated by collision with and ejection of orbital electrons within the food. These collisions create free radicals and ions and bring about physical and chemical changes in the food without raising its temperature. The physical changes include destruction of microorganisms, which are the chief cause of food spoilage. Chemical changes do not usually contribute to the preservation process; indeed, chemical changes are the cause for concern about the wholesomeness of irradiated food and at high radiation doses can cause flavor and odor changes.

The energy level used to treat foodstuffs is never high enough to cause nuclear reactions to take place; hence there is no question of irradiated food becoming radioactive. When checking the wholesomeness of irradiated food, the researcher centers his investigation around possible synthesis of chemical compounds which could harm the consumer if ingested.

Potential Applications

Growth Depression (5 to 15 krads)

Subjecting potatoes and onions to doses of 10 krads inhibits sprouting regardless of storage temperatures; however, since this dose does not inactivate microorganisms, temperature and humidity conditions must be controlled during long-term storage. Storage at 7 to 10°C makes the potatoes suitable for long time periods in the manufacture of potato chips; the lower

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temperatures at which potatoes are usually stored lead to buildup of sugars that cause excessive browning in the final product. This is important to countries that can grow only one crop a year, such as Canada that consequently has to import large quantities of potatoes at high cost from the southern United States.

Effective chemical sprout inhibitors are being perfected that should be economically competitive with radiation. In addition, finish-frying of potatoes with microwaves has recently been suggested as a way to avoid the browning caused by caramelization. There might still be a retail market application for radiation inhibition of sprouting since potatoes are often stored at relatively high temperatures in today's modern apartments. Savings would in any case have to outweigh the process costs of 0.05 cent/lb.

No effective chemical inhibitor of sprouting is available for onions. Irradiation has considerable merit in this case because it can be used to extend the export market. At present, Canadian onions are exported to the West Indies for only 4 months after harvest.

Insect Destruction (10 to 150 krads)

Doses in this range cause sterilization or death to insects infesting food. The Food and Drug Administration (FDA) in the United States has approved this process* for application to wheat and wheat flour. A pilot-plant-scale grain irradiator has been built in Savannah, Ga., and a full-scale grain irradiator† has been constructed in Iskenderun, Turkey. The USAEC plans to petition the FDA for approval of irradiation of mangoes and papayas for this purpose between 1970 and 1972.

Radiopasteurization (100 to 500 krads)

Doses in this range destroy the majority of microorganisms in a food product and effectively extend shelf life under normal handling conditions without causing chemical changes that noticeably affect flavor, odor, or texture of the food.

A dose of 200 krads destroys the molds *Botrytis cinerea* and *Rhizopus stolonifera* on strawberries. This treatment would reduce losses from the present 15% to about 5%. A savings of 4.5 cents/lb would result; processing costs would be about 1 cent/lb. In addition, a shelf-life extension of 4 to 6 days is obtained. A petition to FDA from the USAEC for approval of the irradiation of strawberries is being prepared for submission. Other fruit species that may use this process include sweet cherries, figs, and tomatoes.

Research has shown that 100 to 200 krads will extend the shelf life of many fresh marine products by 2 to 3 weeks at a storage temperature of 0 to 3°C if quality of fish at time of irradiation is high. Products showing promise include pollock, ocean perch, flounder, haddock, sole, halibut, cod, clams, crab meat, lobster, shrimp, oysters, and scallops. Costs of irradiating seafood would be about 0.5 cent/lb for a plant with a throughput of 10,000 lb/hr, providing a dose of 250 krads, and having a source utilization efficiency of 40%. Potential benefits—difficult to establish because they depend on market response to the product—would include expansion of markets inland, ability of the retailer to purchase supplies with more flexibility, use of seafood in processed products unable to be manufactured now because of preservation difficulties, and purification of oysters and clams dredged from polluted waters. Housewives would have a choice—even in places far from the coast—between frozen fish and irradiated fresh fish that would keep in the home refrigerator. Presumably, the trend would be to fresh fish and away from the frozen product.

Radiopasteurization of poultry with 500 krads provides a 2-week extension of shelf life at temperatures of 1 to 4.4°C and effects a 10 and 11 log reduction in *Salmonella* population, thereby eliminating a potential public health hazard. Processing costs are about 0.5 cent/lb; but whether or not this is a sufficient incentive to commercialization, even if health authorities permit it, is not clear. Other products that can be treated to eliminate *Salmonella* include frozen dried eggs, frozen meats, fish meal, and animal feeds.

Most meat is sold in the fresh form, i.e., dressed and chilled, and can be displayed only 3 to 5 days. Because of this perishability, the retailer purchases carcasses, sides, and quarters and performs part of the manufacturing operation at the store (i.e., he converts these large units to retail cuts and packages them). A savings of 2 to 3 cents/lb could be realized if a central cutting facility were used, but this would require a shelf life of 11 to 12 days. To achieve this extension, microbial development must be inhibited and physical and chemical changes in the meat itself must be controlled. A dose of 100 krads will delay microbial spoilage of beef and pork at least 14 days; phosphates effectively control drip, and an oxygen-enriched atmosphere aids in color retention. Alternative

*In February 1969 the Canadian Food and Drug Directorate also approved this process.—JM

†Although the plant was constructed, the Turkish government canceled the project.—*The Editor.*

methods are total freezing and bulk packaging in a controlled nitrogen atmosphere. The cost of radiation processing is about 1.5 to 2 cents/lb.

Radiation-Improved Dehydrated Vegetables (0.3 to 7 Mrads)

Irradiating dehydrated vegetables results in their having shorter rehydration times and increased tenderness. The wide variation in dose—from 0.3 to 7 Mrads—reflects the equally wide biological variation in vegetables. A company using this product would be able to market combinations of vegetables and other dried ingredients that are unheard of now because of the present wide variation in rehydration times. The housewife would have a wider selection of dried soups and stews which would have shorter rehydration times.

Complete Bacteria Destruction (1.5 to 6 Mrads)

Complete sterilization of food after packaging is possible with radiation. The principal food investigated has been meat. A minimum dose of 4.5 Mrads is needed to inactivate *Clostridium botulinum*, but this large dose causes chemical changes in beef, namely, off-odors and off-flavors. These organoleptic changes are believed to be the result of volatile chemical compounds produced by radiation impact on protein and lipid molecules. Irradiation at −30 or −80°C reduces these organoleptic changes. Significant vitamin B destruction has been noticed, but most normal diets provide an excess of this nutrient, and its destruction in meat probably does not present a serious problem. Because the 4.5-Mrad dose is insufficient to destroy the meat enzyme systems, partial precooking is necessary to obtain a shelf-stable product. The cost of radiation sterilization of meat has been estimated at 3 to 5 cents/lb. This particular application received a setback in the United States in April 1968 when the FDA refused to approve radiation sterilization of ham on the grounds that the product had not been proven safe for human consumption.

If radiation-sterilized meats do become available, the housewife will have more refrigerator space available for other items and her choice of a wide variety of heat-and-serve items not presently found in the home.

Summary

The most significant technical difficulty still to be overcome before commercialization of food irradiation becomes a reality is that of obtaining conclusive proof that irradiated food is absolutely safe for consumption. A lesser problem is that of controlling organoleptic changes when the food is treated with high doses of radiation. Costs appear to be justifiable in terms of potential savings. Within the next 5 years, there should be some commercialization of radiation sprout inhibition and radiation pasteurization of some fruits and seafoods. However, in North America and Europe, food irradiation will not be widely commercialized for perhaps 2 decades. Commercialization may well be achieved sooner in some developing countries. Food irradiation does not at this time present a threat to refrigeration. When and if food irradiation does become commercialized, it will be competitive with frozen items in particular. (FEM)

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Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Extracts from a Report on Concrete–Polymer Materials*

Editor's Note: Extracts from the first topical report on concrete–polymer materials are reproduced here because of possible interest to the readers of *Isotopes and Radiation Technology*. This radiation-produced product may have many uses as a new construction material. Because of space limitations, much detailed information and many data have necessarily been omitted, as have most of the 43 figures and 55 tables. For further information on this subject, the interested reader should consult the full report.

Abstract: *Extracts from a joint report [BNL-50134(T-509) and USBR Gen. Rep. 41] describing the preparation, testing, and development of concrete–polymer composites as improved construction materials are given. The studies include the development of methods of preparation of both preformed and premixed concrete–polymer composites. The preparation techniques for preformed concrete include drying, evacuating, monomer soaking, coating, and in situ polymerizing of the monomer either by ^{60}Co gamma radiation or by thermal-catalytic initiation, or by a combination of both. For the premix concrete, either part of the water is replaced with monomer or monomer is added to the fresh concrete mix, and the mix is subsequently radiation or thermally treated.*

Remarkable improvements in properties—including strength, resistance to freeze–thaw cycles, reduced water permeability, and reduced corrosion by distilled water—have been obtained. Examination of the fine structure of the concrete–polymer material by optical and electron microscopy and petrographic techniques is described briefly, and several possible applications of concrete–polymer are mentioned.

The impregnation of concrete with monomer followed by treatment of the monomer with radiation originated with a suggestion by the U. S. Bureau of Reclamation (USBR) of the Department of the Interior to the Division of Isotopes Development of the U. S. Atomic Energy Commission (USAEC) in 1965. The Commission at the time was supporting a program on the development of wood–plastic combinations. The idea was transmitted to the Radiation Division, Nuclear Engineering Department, Brookhaven National Laboratory (BNL) by the USBR and USAEC, and late in 1965 and early in 1966 exploratory experiments were carried out. Mortar bar specimens were impregnated with several gaseous- and liquid-phase monomers and polymerized in situ with ^{60}Co gamma radiation. Results of these initial exploratory experiments, completed early in 1967, showed that complete penetration through the cross section of 1- by 1-in. bars could be obtained with maximum loadings up to 6.6 wt.%. Of the monomers used, ethylene, as gas or dissolved in liquid SO_2, resulted in dispersion of a white powdery material through the cross section of the bar, whereas methyl methacrylate and styrene gave a hard glassy polymer. The USBR measured some of the properties of these initial mortar bars impregnated with methyl methacrylate (MMA) and styrene (S); the main findings were (1) the compressive strength was increased by a factor of 2 to 2.4, (2) the absorption of water was decreased 80 to 98%, and (3) the hardness was increased by a factor of 1.4 to 1.9.

Based on these rather striking initial results, the USBR became interested in the composite material for improving the resistance of concrete to sulfate, hot brine, and distilled water and to freeze–thaw and cavitation conditions. Of particular interest was the possible application of the composite material as a construction material for water-desalination plants in

*M. Steinberg, L. E. Kukacka, P. Colombo, J. J. Kelsch, B. Manowitz, J. T. Dikeou, J. E. Backstrom, and S. Rubenstein, Concrete-Polymer Materials, First Topical Report, BNL-50134(T-509)—USBR Gen. Rep. 41, Joint Report of Brookhaven National Laboratory (USAEC) and U. S. Bureau of Reclamation (U. S. Department of the Interior), December 1968.

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which most of these conditions exist. The Office of Saline Water (OSW) was and is currently supporting a program at the USBR to determine the behavior and resistance to deterioration of concrete exposed to hot brine and distilled water. Concurrently, a program to develop and evaluate protective coating materials for application to concrete surfaces is in progress; however, a satisfactory coating has not yet been found that would adequately protect concrete from hot brine at temperatures ≥290°F (≥143°C).[1]

Another potential large-scale application is in concrete pipe. A preliminary cost estimate including a conceptual design of a plant to produce a concrete–polymer unreinforced drain pipe was prepared by BNL.[2] This analysis indicated that concrete–polymer material might possibly compete with protectively coated concrete and glazed pipe. As a result, a joint program supported by the USAEC, USBR, and OSW was established between the USBR and BNL.

The long-range objectives of the joint program are the investigation and development of a concrete–polymer composite for application as a new and improved construction material. The program includes the development of techniques for the preparation of the concrete–polymer material, the measurement of pertinent physical and chemical properties, the preparation of full-scale concrete products, and the development of conceptual process designs for, and evaluation of, various specific applications. Generally, BNL investigates the preparation techniques, impregnates and polymerizes test specimens provided by the USBR, and develops conceptual process designs for formation of concrete–polymer products; the USBR performs the physical property measurements on test specimens, including pertinent corrosion tests. A basic research effort is maintained at both laboratories to help understand the fundamental nature of concrete–polymer composites. Included in the program is polymerization by thermal-catalytic means as well as by radiation initiation. In addition to treating preformed-concrete specimens, the investigations include adding monomers to fresh mix concrete and subsequently polymerizing, either before or after allowing the concrete to harden. This technique was initially investigated at BNL and is now being pursued by the USBR.

Essentially, one mortar-mix series and four different concrete-mix series have been used in investigations to date. Ingredients, methods of fabrication, and methods of curing have varied for each series and are discussed in detail in the full report; basic information can be found in the *U. S. Bureau of Reclamation Concrete Manual.*[3]

Concrete Formation

Portland cement concrete is composed of sand, gravel, crushed rock, or other aggregates, held together by a hardened paste of hydraulic cement and water. The thoroughly mixed ingredients, when properly proportioned, make a plastic mass which can be cast and molded into a predetermined size and shape. Upon hydration of the cement by the water, portland cement concrete becomes stonelike in strength and hardness. With the addition of reinforcement to supply needed tensile strength, concrete has become the foremost structural material.

A relatively low cost and the capacity to be formed into a wide variety of sizes and shapes are among the major advantages in the use of portland cement concrete. Although concrete is an excellent building material, certain limitations in its use must be recognized. Its most objectionable characteristic is volume change. Internal stresses developed within concrete resulting from expansion and contraction due to temperature and moisture changes, combined with its relatively low and slow-developing tensile strength (~10% of its compressive strength), make it prone to cracking.

In addition to strength and thermal properties, other properties of concrete that can be affected by its composition and should be considered in its use are its resistance to chemical attack, physical deterioration due to weathering (particularly freezing and thawing), permeability and absorption, creep, and abrasion and cavitation resistance.

Included in the full report is a list of definitions of some of the more pertinent terms related to concrete technology, which are principally from the American Concrete Institute's *Cement and Concrete Terminology.*[4]

Premix Series

A series of mixes was made at BNL to determine the feasibility of incorporating monomers integrally into the fresh concrete mixture.

Radiation-Initiated Polymerization. Two control mixes (no monomer) and five test mixes using various monomers were made for radiation-initiated polymerization. Mixes are identified as follows.

1. Control concrete containing water with no monomer.
2. Concrete containing 8 vol.% styrene.
3. Concrete containing 8 vol.% methyl methacrylate.
4. Concrete containing 8 vol.% acrylonitrile.

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5. Concrete containing 8 vol.% of a mixture of styrene and 1% divinyl benzene.

6. Control concrete containing methyl alcohol as a substitute for water.

7. Concrete containing 8 vol.% styrene and methyl alcohol as a substitute for water.

The control mix with water (mix 1) was made by using aggregate graded to ¾-in. maximum size, 5.8 sacks of cement/cu yd, and a 0.51 water/cement ratio. In mixes containing 8 vol.% liquid monomer (mixes 2 to 5), a 1 : 1 volume replacement of water with monomer resulted in mixes that were too dry.

For mixes with workability comparable to that of the control, it was necessary to substitute monomer for both water and cement. Eight vol.% liquid monomer was best incorporated into the mix by reducing the water content of the control mix by 25% and the cement content by 20%, and adjusting the sand and coarse aggregate accordingly to produce total yield quantities similar to those of the control. The water/cement ratio in mixes 2 to 5 was 0.48.

Substituting methyl alcohol for water results in a wetter concrete mix having considerably more slump. For producing a mix having a consistency similar to that of mix 1 (control mix made with water), mix 6 (methyl alcohol substituted for water) was made with a cement content similar to that of mix 1, but the alcohol/cement ratio was reduced to 0.37. Mix 7, made with methyl alcohol substituted for water and containing 8 vol.% styrene, required adjustments in relation to its control (mix 6) similar to those made for mixes containing water and 8 vol.% monomer. This resulted in an alcohol/cement ratio of 0.35 for mix 7.

Polymerization of specimens cast from these concretes was performed either immediately after their fabrication or the next day, after hardening. Following removal of molds from specimens 1 day old, specimens were cured for 27 additional days, then shipped to the USBR for testing. Specimens made with water were cured by storage in water. Similarly, those made with methyl alcohol were cured by storage in methyl alcohol.

Thermal-Catalytically Initiated Polymerization. One control and four test mixes in which methyl methacrylate was used were made for thermal-catalytically initiated polymerization. Mixes are identified as follows.

1. Concrete containing 1 wt.% of a mixture of methyl methacrylate and 5% benzoyl peroxide, for immediate polymerization.

2. Concrete as in item 1, but for polymerization after hardening.

3. Control concrete (no monomer), for thermal treatment after hardening.

4. Concrete containing 16.4 wt.% of a mixture of 40% polymethyl methacrylate and 60% methyl methacrylate with 5% benzoyl peroxide, for immediate polymerization.

5. Concrete as in item 4, but for polymerization after hardening.

Concrete used for these experiments was ready-mixed concrete containing ¾-in. maximum size aggregate. Information was not available on cement content, water/cement ratio, air entrainment, etc., for this concrete. Following removal of molds from specimens 1 day old, specimens were cured by storage in water for 27 additional days, then shipped to the USBR for testing.

Preformed-Concrete Series

Following evaluation of results of the mortar-mix series, experiments were undertaken to develop techniques of in situ impregnation and polymerization of monomers in hardened concrete and to determine properties of resultant polymer-impregnated concrete.

Concrete used in these investigations contained Clear Creek aggregate graded to ¾-in. maximum size, 5.5 sacks/cu yd of a type II laboratory blend cement (M-5500), 0.51 water/cement ratio, and air entrainment.

In the initial mixes, sand passing through a 200-mesh sieve was used, leaving a slightly harsher mix that would be more porous and more amenable to a greater degree of impregnation by monomer. Subsequent tests of this concrete indicated that its absorption and permeability characteristics were not greater than those normally expected for similar concrete containing the minus 200-mesh-sieve-sized sand. In view of this, and in order to have test data on a concrete similar to that which would be produced commercially, the minus 200 sieve-sized sand fraction was included in later mixes.

To obtain comparative data on impregnation and polymerization techniques and on resultant properties of polymer-impregnated concrete, similar concrete-mix proportions were used for all specimens in the initial survey tests and in the subsequent tests for thorough evaluation of monomers. Thorough evaluation tests for a given monomer were performed on specimens cast from a single mix. The mix was duplicated for each

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monomer to be evaluated and for evaluation of control (no monomer) concretes.

Immediately after fabrication, all specimens in the initial survey tests and in tests for thorough evaluation of monomers were fog-cured for 28 days. Testing of initial survey test specimens was not initiated at a uniform time after fabrication, the tests being performed as impregnated specimens became available. However, for thorough evaluation of monomers, tests were initiated 120 days after fabrication of specimens.

Investigations also are scheduled to determine the effect of impregnation and polymerization of monomers in concretes of varying compositions. The first concrete selected for these investigations was a high-strength concrete used by the USBR in connection with tests performed for the OSW to determine the corrosion resistance of concrete to brine and distilled water at elevated temperatures and pressures.

Specimens for these tests were fog-cured for 28 days after fabrication, then stored in laboratory air for ~3½ years, until shipped to BNL. Their age at time of testing was 3 years and 8 months.

Polymer Formation

This section deals with the selection and polymerization of monomers by both radiation and thermal methods for application to concrete–polymer formation. The techniques of thermal polymerization are drawn from many years of experience,[5] and those for radiation polymerization over the past two decades.[6,7] The application to preparation of composites is relatively recent and has been concerned mainly with wood–plastic combinations.[8] There is little precedent for preparation of plastic-impregnated concrete or a concrete–polymer material.

Criterions for Monomer Selection

A number of factors must be taken into account in selecting a monomer for application in a concrete–polymer material. These can generally be categorized under (1) cost of monomer, (2) processability and cost of processing the monomer, and (3) properties of the polymer and its combination with concrete. Several of the leading candidate monomers for the present research and application development program are listed in Table 1.

When dealing with concrete and concrete–polymer as a construction material, cost is a most important consideration. Concrete can be produced at a raw material cost of about $10 to $12/cu yd in the United States. This is equivalent to ~0.25 cent/lb. In the case of plastics, the least expensive organic monomer, ethylene, costs a minimum of 2 cents/lb, while the

Table 1 Survey of Monomer Systems for Concrete Impregnation

	Polymer softening point, °F (°C)	Compressive strength, psi	Tensile strength, psi	Radiation dose for 100% conversion at 5.3×10^5 rads/hr, Mrad	Cure time in oven at 167°F (75°C), hr; monomer +1% BzP*	Monomer cost, $/lb
Methyl methacrylate	212 (100)	14,500	9,500	4.8	1.25	0.21
Styrene	230 (110)	13,500	8,000	50.0	8.0	0.09
Acrylonitrile	518 (270)	–	–	1.6	0.6	0.145
Styrene–acrylonitrile, 60/40	230 (110)	15,500	10,500	5.1	1.0	0.115
Vinyl chloride [vapor pressure, 30 psig at 68°F (20°C)]	176 (80)	10,500	7,000	–	–	0.082
Vinyl acetate	158 (70)	–	<5,000	5.1	1.0	0.14
Isobornyl methacrylate	297 (147)	–	–	1.2	0.75	(0.50–1.00)
Trimethylolpropane trimethacrylate	–	–	–	0.9	0.5	0.98
Diallyl phthalate	311 (155)†	22,000	6,500	58.0	8.0	–
Plaskon-941‡ (polyester–styrene)	195 (92)†	22,500	10,600	0.5	0.75	0.22
Hetron-197§ (polyester–styrene)	311 (155)†	24,000	16,200	1.2	0.75	0.445
Epocryl U-16¶ (epoxy–styrene)	300 (148)†	25,000	10,200	5.1	1.5	0.40

*Benzoyl peroxide.
†Heat deflection temperature, ASTM D648.
‡Allied Chemical Corp.
§Hooker Chemical Corp., Durez Plastic Division.
¶Shell Chemical Co.

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more common liquid monomers range from 9 cents to 40 cents/lb, and specialty monomers are >$1.00/lb. Thus additions of monomer to concrete can significantly increase the cost of a concrete product on a unit-weight basis. However, an important balancing factor in bulk air-entrained concrete is that its void fraction is limited to ~13 vol.%. This is equivalent to ~6 wt.% of unit density polymer when the void spaces in the concrete are filled with polymer. Thus, by a relatively small addition of monomer, concrete can be fully impregnated and relatively large changes in its properties could result. This is quite different from impregnation of other materials such as wood, in which the void fraction in the cellular structure can be as high as 60% and the polymer content of the composite is consequently high. Because of this, the cost of wood–plastic is markedly higher than that for wood alone. However, even with a 6% weight loading in concrete, with a monomer cost of 20 cents/lb, 1.2 cents/lb is added to 0.25 cent/lb concrete, or a fivefold increase in cost per unit weight of concrete. This increase appears to preclude the use of concrete–polymer material except for very special applications, such as those involving highly corrosion-resistant conditions or aesthetic values. If, however, the cost of this composite material is related to the volume of a finished product or a cubic yard in place cost, its use is entirely justifiable. For example, the cost of a finished concrete product (e.g., pipe) is ~$120/cu yd of concrete. The cost of conventional structural concrete in place is not much different from this, namely, about $135/cu yd. It is estimated that the production by radiation polymerization of a finished concrete–polymer product would roughly double the cost of concrete.[9] The cost of concrete–polymer would then total $240/cu yd. This applies for a monomer cost of $0.20/lb, which at 6% loading would amount to $48/cu yd of concrete. This treated concrete, based on measurements made here, can have properties such as compressive and tensile strength as much as four times as great as those of conventional concrete. This great increase would permit the designer to reduce the section of structural members to ¼ that of the conventional concrete section. As a consequence, a conventional concrete member costing $120 could be replaced with impregnated concrete for $240/4, or $60. Assuming the polymerization cost to remain roughly the same at $72/cu yd of concrete–polymer ($240 total, less $120 for concrete, less $48 for monomer), a monomer cost of $1.20/lb, or, at 6% loading, monomer at ~7 cents/lb of concrete, could be justified as just breaking even with that of conventional concrete. Thus the application of the composite material could be rather wide, and some of the more exotic and expensive monomers could be considered. To date, only the very-low-cost common ones, such as methyl methacrylate, styrene, acrylonitrile, vinyl acetate, vinyl chloride, and ethylene, have been considered.

From a process point of view, the monomer should have a low enough viscosity to readily penetrate the microporous structure of the concrete. The boiling point of the monomer is important in determining whether a closed vessel and pressure are needed to contain the monomer surrounding the concrete piece to be impregnated. Methyl methacrylate, styrene, acrylonitrile, and vinyl acetate (liquid monomers) have low viscosities and boiling points above 158°F (70°C), so that impregnation can take place at atmospheric pressure and ambient temperature. Vinyl chloride requires pressure in an enclosed system; ethylene also requires pressure to obtain any appreciable mass loadings and high polymerization rates. The radiation dose required for polymerization and the thermal treatment time with peroxide catalyst also affect the economics of the process. In the case of styrene, however, larger doses of radiation and long curing times for thermal-catalytic treatment are required, as compared with other monomers such as methyl methacrylate and acrylonitrile.

Monomers must also be selected on the basis of the properties of the resultant polymers and the application for which they are intended. For use of concrete as a construction material for the multistage flash distillation units in water-desalination plants, a continuous hard glassy polymer coating providing complete coverage of the surface and capable of withstanding hot brine, vapor, and distilled water at temperatures between 101°F (38°C) and 290°F (143°C) is required. Because the polymers of methyl methacrylate, styrene, vinyl acetate, and polyethylene have softening points below the maximum value, their use would be precluded throughout a portion of this temperature range. This does not necessarily mean that these polymers will flow from the concrete microporous structure at temperatures above their softening point. However, for materials with softening points above 290°F (143°C), there would be a greater degree of certainty that the polymer would remain fixed. Acrylonitrile with a high softening point [518°F (270°C)] normally forms a white powder that is insoluble in its monomer. However, a white powdery impregnation was not readily observed with acrylonitrile, as it was with ethylene, and a coherent impregna-

tion may yet be possible with acrylonitrile. Styrene is added to acrylonitrile in a copolymer system to produce a continuous hard coating material; however, the softening point is reduced to 230°F (110°C). It is possible to add a cross-linking agent to a monomer to increase its thermal resistance and stability. Trimethylolpropane trimethacrylate (TMPTMA) is a suitable cross-linking agent and has been used with methyl methacrylate to increase its softening point to >392°F (>200°C) without distortion. This is an excellent candidate for the higher temperature OSW application.

Another possibility is the use of thermosetting resins such as polyesters and epoxies. Two unsaturated polyesters, Plaskon-941 and Hetron-197, are both mixed with styrene for polymerization. In the case of Hetron, only partial penetration could be obtained because of the increased viscosity. Dilution with styrene reduces the viscosity and increases the penetrability; however, the heat-distortion temperature is then lowered. Epocryl, an epoxy–styrene, has good heat-distortion resistance. The radiation dose and thermal curing requirements are relatively low for the thermosetting resins.

Isobornyl methacrylate shows some promise as a vinyl monomer that would yield a polymer with a high softening point. However, it is a new monomer, currently limited in availability and relatively high in cost.

The compressive and tensile strengths listed in Table 1 show the relatively higher strengths of the thermosetting resins as compared with the vinyl polymers. This could be reflected in increased strength of the resultant concrete–polymer materials. Most of the polymers listed in Table 1 can withstand the usual outdoor weathering conditions as well as mildly corrosive acidic and alkaline conditions.

In addition to the monomers in Table 1, ethylene, vinylidene chloride, and α-chlorostyrene are of interest in the present program.

The method of approach in surveying a selected monomer is to (1) survey the literature and obtain recommendations from polymer chemists and monomer manufacturers, (2) determine the radiation and thermal requirements for polymerization of monomer, (3) measure selected properties of polymer such as softening point, (4) develop techniques for impregnation of concrete, (5) prepare 3-in.-diameter by 6-in.-long and 6-in.-diameter by 12-in.-long cylindrical concrete specimens by both radiation and thermocatalytic means, and (6) measure polymer loading and several physical and elastic properties germane to the proposed exposure and use.

Requirements for Radiation Polymerization

The use of ionizing radiation as a method for initiating addition polymerizations is well known. Perhaps the most important single factor in favor of radiation-induced polymerization is the ability of radiation to initiate chain reactions at low temperatures.

Since radiation polymerization is known to proceed by the formation of free radicals, the monomers selected as possible candidates for concrete impregnation were mainly of the type known to propagate by a free-radical mechanism. For impregnation of concrete it is more convenient to use a free-radical polymerization mechanism than an ionic mechanism, since the former is less sensitive to impurities and temperature variations. In addition, in a free-radical mechanism, high positive activation energies are obtained as compared with the low or negative values obtained with an ionic mechanism. Thus the polymerization rates can be increased by raising the reaction temperature or by using the heat of reaction. Studies on the gamma-ray-initiated polymerization of styrene and methyl methacrylate at various temperatures showed overall activation energies of 7.15 and 4.9 kcal/mole, respectively, for the two monomers.[10]

Under constant radiation and temperature conditions, the rate of polymerization varies for different monomers. This is governed by the rate constant for the propagation step.

Another parameter affecting the polymerization rate is radiation intensity, usually given in rads/hr. For low radiation intensities a classical expression that has been experimentally confirmed for pure monomers states that the reaction rate is dependent on the square root of the intensity.[6,7] The rate of polymerization can therefore be increased by increasing the radiation intensity. The total dose requirement, however, also increases with increasing radiation intensity. At a very high radiation intensity, or at high rates of radical initiation, the polymerization rate reaches a value that does not increase further with increasing radiation intensity. These values can vary, depending on the monomer used and on the efficiency of radical production.

The radiation doses required to give 100% conversion of several monomers under study have been experimentally determined. The data, obtained at a radiation intensity of 5.3×10^5 rads/hr and at 68°F (20°C), are given in Table 1. Estimates of the total dose requirement and overall polymerization rate at an intensity of 10^5 rads/hr have been calculated from the

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experimental data, assuming a square-root dependence on rate. These estimates are summarized in Table 2.

Requirements for Thermal-Catalytic Polymerization

Most monomers that can be polymerized by free radicals also polymerize at elevated temperatures. In performing thermal polymerization studies, certain temperature limitations must be observed and reaction temperatures must remain below the boiling point of the monomer used. However, in most cases the polymerization rates are extremely low. To increase the rate of radical production, free-radical-type catalysts are added to the systems. The catalyst chosen for this study was benzoyl peroxide, because it is the most commonly used peroxide and also because of its stability at room temperature.

A study of the thermal-catalytic method involves the effect of catalyst concentration and temperature on the polymerization rate. To date, quantitative data have been obtained for styrene, methyl methacrylate, and isobornyl methacrylate. The experiments were conducted by dissolving catalysts of different concentrations in the respective monomers. Samples about 10 ml in volume were exposed to varied temperatures for the time required to give 100% conversion.

It has been observed from experimental data that, in most cases, addition of >1 to 3% peroxide does not appreciably decrease the polymerization time. This phenomenon can probably be explained in terms of the decomposition mechanism for benzoyl peroxide. When the peroxide molecules are heated, bonds break and radicals are formed. The peroxide molecules are subject to decomposition by several different homolytic mechanisms.

Peroxide decomposition reactions also can lead to chain branching reactions, and explosions may result. This makes all peroxide catalysts dangerous to handle. Jarring or heating can initiate explosions in peroxides. Special precautions must therefore be taken when

Table 2 Effect of Radiation Intensity on Polymerization Time for Various Monomers

Monomer	Radiation intensity, 10^5 rads/hr	Overall rate of polymerization, % conversion/hr*	Radiation time for 100% conversion, hr	Total dose, 10^6 rads
Methyl methacrylate	5.3	11.0	9.1	4.8
	1.0	4.8	21.0	2.1
Styrene	5.3	1.06	94.3	50.0
	1.0	0.5	217.0	21.7
Acrylonitrile	5.3	33	3.0	1.6
	1.0	14.5	6.9	0.7
Styrene–acrylonitrile	5.3	10.4	9.6	5.1
	1.0	4.5	22.0	2.2
Vinyl acetate	5.3	10.4	9.6	5.1
	1.0	4.5	22.0	2.2
Isobornyl methacrylate	5.3	44.0	2.3	1.2
	1.0	18.9	5.3	0.5
Trimethylolpropane trimethacrylate	5.3	58.8	1.7	0.9
	1.0	25.6	3.9	0.4
Diallyl phthalate	5.3	0.91	105.6	58.0
	1.0	0.4	243.0	24.3
Plaskon-941 (polyester–styrene)	5.3	106.0	0.9	0.5
	1.0	47.6	2.1	0.2
Hetron-197 (polyester–styrene)	5.3	44.0	2.3	1.2
	1.0	18.9	5.3	0.5
Epocryl U-16 (epoxy–styrene)	5.3	10.4	9.6	5.1
	1.0	4.6	22.0	2.2

*On the basis of experimental data given in Table 1 for an intensity of 5.3×10^5 rads/hr, values at 1×10^5 rads/hr were calculated. Assuming square-root intensity dependence, overall rate = 100%/time for 100% conversion of monomer to polymer.

dealing with large amounts of monomer–peroxide mixtures.

Additives

Various types of additives can be used in a polymerization system to increase or decrease the polymerization rate or to improve the polymer properties. The main objective in investigating the use of additives is to reduce the radiation polymerization requirements for the concrete impregnation work to values below those obtained with some of the monomers alone.

Inhibitor. Since most monomers are unstable under ambient conditions, an inhibitor is added to the monomer by the manufacturer prior to shipping. The inhibitor acts as a radical suppressor or scavenger. When the monomer is exposed to radiation or heat and catalyst, the radicals formed initially must overcome the inhibitory effect by reaction with the inhibitor, or direct radiation decomposition of the inhibitor must occur, before a chain reaction between monomer molecules can take place. This results in a "lag time," or induction period, before polymer can be formed. The inhibitor can be removed from the monomer by distillation or by washing with a dilute caustic solution. Aside from being a costly operation, the removal of inhibitor presents a safety hazard, particularly in a large-scale operation. Sometimes the amount of inhibitor is increased to give an extra margin of safety or to prolong the shelf life of the monomer.

Catalyst. As shown in Fig. 1, the induction dose required to overcome the inhibitor was ~3 Mrads. To eliminate this induction period and possibly increase the rate of polymerization in a radiation field, ⩾1% benzoyl peroxide was added to the monomer; the induction period was eliminated and the polymerization rate increased.

The disadvantage of using this technique for large-scale operations is the potential danger involved in handling large quantities of monomer to which peroxide catalyst has been added.

Cross-Linking Agent. The advantage of incorporating a cross-linking agent in a polymerization system lies primarily in improvement of the thermal and chemical resistance properties of the polymer. As mentioned earlier, an efficient cross-linking agent used in this work was TMPTMA. This monomer is capable of homopolymerizing by itself to form a completely three-dimensional cross-linked polymer; it can also be incorporated in varying concentrations with other

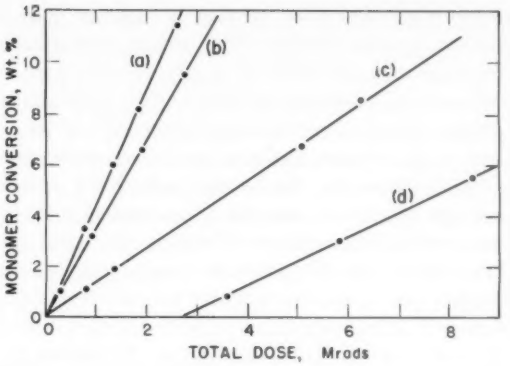

Fig. 1 Radiation-induced polymerization of styrene with (*a*) 3% benzoyl peroxide, (*b*) 1% benzoyl peroxide, (*c*) inhibitor removed, and (*d*) inhibitor present. Radiation intensity 2.6 × 10^5 rads/hr, temperature 68°F (20°C).

polymers to give cross-linked materials. By using this material, a cheaper monomer can be upgraded and given high thermal stability as well as other desirable properties, such as inertness to solvents.

Table 3 gives some results of using TMPTMA with styrene and methyl methacrylate monomer. The data indicate that the addition of TMPTMA to the monomers greatly reduced the radiation dose required to obtain 100% conversion. In addition, softening points for the cross-linked styrene and methyl methacrylate increased from ~212 to 392°F (~100 to 200°C).

Wetting Agent. Wetting agents can effectively be used to increase the rate of penetration of monomer through a porous material. For concrete impregnation work, particularly when complete loading is desired, the use of wetting agents may not be desirable. Wetting agents are composed, for the most part, of alcohols. If alcohols are incorporated into the monomer, a fraction of the loading in concrete will consist of alcohol. After polymerization, the alcohol will be locked into the concrete–polymer network, occupying some of the void space in the concrete. Possibly the most detrimental effect is that alcohols act as precipitants for polymers. Therefore, as polymerization takes place, the alcohol precipitates the polymer out of the monomer–polymer solution. The precipitated polymer is usually in the form of a fine powder. This will destroy the continuous, glassy, hard polymer structure desired in most cases.

Promoter. Promoters are used to reduce the temperatures at which catalysts normally decompose. In the case of the radiation process, promoters have little

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value since they have nothing to act on. For the thermal process, however, they can be very effective. Normally the half-life of a 1% benzoyl peroxide solution is ~60,000 hr at 68°F (20°C). Addition of a promoter (e.g., dimethylaniline) in the same concentration range reduces the half-life of the peroxide to ~10 min. However, this is less practical for concrete impregnation work, since the entire mass of monomer used for soaking is likely to polymerize shortly after introduction of the promoter in any impregnation vessel.

in the specimen and its effect on the overall system must be known. Serious consideration must be given to (1) the miscibility or solubility of a particular monomer in water, (2) displacement of water by monomer, (3) internal resistance or blocking by water, (4) monomer density, (5) effect of water on polymerization rates, and (6) effect on polymer properties. A study of these effects will be made in the future. In the present work the main effort has been toward obtaining a maximum monomer loading in a concrete specimen. To achieve this, the most important factor is the

Table 3 Effect of TMPTMA* on Dose Requirements and Polymer Properties

Monomer	Dose required for 100% conversion,† 10^6 rads	Remarks
Styrene	50.0	Soluble (noncross-linked)
TMPTMA	0.9	Cross-linked
Styrene + 10% TMPTMA	3.9	Cross-linked
Styrene + 20% TMPTMA	3.8	Cross-linked
Methyl methacrylate	4.8	Soluble (noncross-linked)
Methyl methacrylate + 10% TMPTMA	1.6	Cross-linked

*Trimethylolpropane trimethacrylate.
†Radiation intensity, 5.3×10^5 rads/hr.

Concrete–Polymer Preparation Techniques

Concrete Preparation and Impregnation Techniques

For determination of the conditions required to obtain any desired degree of monomer loading in a concrete specimen, the various parameters that alone or in combination give optimum preparatory procedures must be investigated. The effects of several of these parameters on monomer loading are being studied.

Drying. Normally a small fraction of the total weight of a concrete specimen consists of water. This can be due to free or unreacted water remaining during the hydration cycle, absorption of moisture during the curing period at 100% humidity, or other postcuring environmental factors. Hence the amount of water contained in concrete can vary from batch to batch. The amount of monomer that can be loaded into a concrete specimen is limited by the amount of free water occupying the total void space. To quantitatively determine the rate of monomer penetration through a concrete specimen of a given thickness and under a given set of conditions, the concentration of free water removal of free water from the specimen. This can be done by vacuum drying or by thermal drying.

Vacuum Drying. Several 3- by 6-in. cylindrical concrete specimens were placed in a small vacuum chamber that was evacuated at 68°F (20°C) to ~3 in. Hg. A specimen was periodically removed from the vacuum chamber and weighed, and the weight loss was recorded as a function of vacuum time. The samples usually contained 3.4 wt.% of free water. With this technique the rate of water removal over a 96-hr period was 0.007%/hr for a weight loss, based on total weight of specimen, of only 0.7%.

Thermal Drying. The time required to thermally dry concrete specimens was also measured. For this work a large-capacity, forced-draft type of oven was used. Since this method seemed more practical than the vacuum method, drying curves were obtained at temperatures of 221°F (105°C), 257°F (125°C), and 302°F (150°C). The data indicated that at 221°F a drying time of ~100 hr is required to obtain equilibrium weight and that at 302°F the drying time required to expel all free water from the samples is 8 hr, compared with 20 hr at 257°F.

Evacuating. Another important parameter to consider in obtaining a maximum loading or improving the efficiency of monomer penetration through a concrete specimen is evacuation of the specimen prior to monomer soaking. The technique used to load a concrete specimen with monomer is to flood the contained specimen with the monomer while maintaining ~1 in. of monomer liquid around the specimen. When an air-saturated specimen is placed in this environment, some of the surface and near-surface air is displaced by the monomer. However, as the monomer penetrates the specimen in all directions, the entrapped air becomes compressed toward the center of the specimen. When the internal and external pressures equalize, the rate of monomer penetration diminishes and total or maximum monomer loading cannot take place. Evacuation down to <~3 in. Hg pressure was usually used. A maximum possible loading value of ~6.7 wt.% has been established for this work, based on information on void volume from the Bureau of Reclamation and on other tests for this type of concrete.

Soaking and Diffusion of Monomer Through Concrete. The technique at present is to soak a concrete specimen by completely surrounding it with monomer. Soaking is measured in terms of the time required for a monomer to uniformly penetrate a given thickness of concrete per unit surface area exposed to the monomer. The time required for a monomer to give any desired degree of penetration and loading depends on (1) monomer viscosity, (2) monomer wetting properties, (3) preparation of specimen prior to soaking, and (4) type of concrete.

For this study, methyl methacrylate monomer was used. The monomer contained a red dye to allow visual determination of the degree of monomer penetration. Monomer loading was determined by weight increase. Before soaking, each sample was dried to equilibrium weight and evacuated for 2 hr. The specimens consisted of 3-in.-diameter by 6-in.-long cylinders of type II cement with air entrainment and a maximum aggregate size of ¾ in. Under the above conditions, complete penetration of monomer occurs within 30 min. At this point the monomer loading is 3 wt.%; it increases with soak time to a maximum of ~6.5% at 5 hr. Since 3-in.-diameter specimens were used for this work, the measurements were limited to penetration depths of 1½ in.

For expanding this range the following technique was devised. A 3-in.-diameter by 6-in.-long cylinder of concrete was dried to equilibrium weight. The outer surface, except for the bottom and top, was coated with epoxy to make it impermeable to the monomer. The specimen was placed in a 4-in.-diameter container with a spacing of ~½ in. between its bottom and the container. The vessel was then filled with methyl methacrylate monomer containing a red dye to a level slightly below the upper surface of the specimen. In this way monomer penetration could be measured through a 6-in. depth. The initial differential pressure across the specimen was 5 in. of liquid methyl methacrylate. At varying times the specimens were removed from the medium and halved longitudinally. The depth of penetration as indicated by the red dye was measured and plotted as a function of time. This experiment was conducted under atmospheric conditions. The initial rate was 0.5 in./hr at 1 in. concrete penetration, decreasing to about 0.18 in./hr after 5 in. penetration. Further studies of this type will be performed using vacuum and pressure conditions to determine the rate of diffusion of monomer through concrete under varying conditions.

Pressurizing. In a further attempt to reduce the time required for monomer loading in a concrete specimen, pressure was applied as the last step in the procedure given above. After covering the concrete specimen with monomer, constant pressure was maintained over the monomer for the duration of the loading cycle. For this work, 5 psig of nitrogen gas was used. Data show that for nonevacuated specimens the use of pressure has little effect on monomer loading. A most significant increase in loading was observed, however, when pressure was applied to specimens evacuated prior to soaking.

Coating Techniques

The materials thus far considered for use in impregnating concrete can be generally classified as either low-viscosity or relatively high-viscosity materials. The low-viscosity materials are monomers such as styrene and methyl methacrylate. A much higher viscosity is characteristic of polyesters, epoxies, and other monomer–polymer combinations. Since the initial interest was in complete penetration of monomer throughout a concrete specimen, the most likely candidates were the low-viscosity materials. Although these materials gave maximum loadings in relatively short times, the outer surfaces of the specimens did not have a continuous polymer coating after treatment. This was due to monomer evaporation during treatment and, most importantly, to the relatively nonabsorbing properties of the aggregate and other materials

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exposed on the surface of the concrete. This could be a problem in certain applications for which exposure of concrete to a corrosive environment is undesirable. One solution to this problem is to coat the monomer-loaded specimen with a monomer–polymer mixture.

Dipping. In preparing a monomer–polymer solution for dipping or coating a monomer-loaded specimen, certain criterions must be met.

1. The viscosity of the monomer–polymer mixture should be such that all voids on the outer surfaces are filled and the mixture does not "run" off the surface of the specimen before the polymerization is completed.

2. The monomer–polymer mixture should be of the same unit structure as the monomer in the saturated specimen. For example, if a concrete specimen is loaded with styrene monomer, the monomer–polymer mixture should consist of polystyrene dissolved in styrene. This is important, since the depth of penetration of a monomer–polymer mixture into a previously loaded specimen is only ~1/16 to 1/8 in. By using the same materials, a compatible homogeneous product results, with a molecular interlock between the outer and inner surfaces. If the monomer–polymer mixture has a structure other than that of the loaded monomer, a heterogeneous polymerization may occur, and the result may be a surface skin that will eventually peel or become detached from the concrete surface. Other complications such as incompatible hardening times and differences in expansion coefficients can also result in a poor surface coating.

Although the dipping method was effective in the case of radiation polymerization at room temperature, the problem of producing a continuous surface coating was more acute with thermal polymerization. To minimize evaporation, the specimens were wrapped in 0.5-mil polyethylene film immediately after loading with monomer and prior to polymerization.

It may also be possible to obtain a sufficiently protective coating of the surface concrete without a dipping technique. As long as the concrete and aggregate surfaces are wetted by monomer, an adherent polymer coating could result. The proof of this must necessarily be the performance of specimens in experimental tests.

Wrapping. Two 3-in.-diameter by 6-in.-long specimens were prepared and loaded to ~6.5 wt.% with methyl methacrylate monomer. Immediately after soaking, one specimen was wrapped with polyethylene. Both samples were allowed to stand at room temperature. Periodically the samples were weighed and the weight loss was plotted as a function of time. The object of the experiment was to simulate monomer loss during the residence time of a specimen in a radiation field at 68°F (20°C). For an unwrapped sample the evaporation was quite rapid, with a monomer loss of ~50% of the initial loading within 6 hr. In comparison, the wrapped sample lost only 0.5% of the total monomer within 1 hr and then remained constant.

The same experiment was performed with specimens that had been dipped after soaking into a monomer–polymer mixture containing 10% polymethyl methacrylate. In both cases the weight loss for the unwrapped specimens was essentially the same, regardless of dipping. For the wrapped specimens, dipping further minimized monomer loss.

Although this work was not extended to thermal conditions, it is obvious that, at least for the unwrapped specimens, the monomer loss would be much greater than that obtained at 68°F (20°C).

For applications in which only a surface coating is desired, it is more convenient to use the higher viscosity materials. The viscosities of styrenated polyesters and styrenated epoxies are such that a continuous coating can easily be applied to a concrete specimen by soaking, dipping, or spray or brush application. The important consideration in any of these applications is the depth of penetration of the material used. A superficial coating may fail because of poor adhesion on the surface of the concrete and contraction of the material during polymerization. A more resistant coating results from in-depth penetration of these materials. At present the best method of obtaining in-depth coatings is simply to soak the concrete in these materials for a time period adequate for the desired penetration. The time required depends on the viscosity of the material used.

Several specimens have been coated in-depth by using the above procedure. The materials used were a styrenated epoxy–acrylate with a viscosity of 22 to 24 centipoises at 77°F (25°C) and a styrenated unsaturated polyester with a viscosity of 800 centipoises at room temperature. The concrete specimens were soaked for 3 hr in these materials, to which a red dye had been added. After soaking, the specimens were wrapped and the monomer was polymerized in a radiation field. The specimens were then cross-sectioned and the degree of penetration was measured. The styrenated epoxy showed a penetration of 1/4 to 1/2 in., and the styrenated polyester with the higher viscosity gave a penetration of 1/16 to 1/8 in. Both specimens had an impermeable and continuous surface coating. Styrenated polyesters and epoxies are desira-

ble for coating applications because of their high strength, high-temperature resistance (due to a large extent to the three-dimensional cross-linked network formed), and the fact that the viscosity of the base mixtures can be varied by adding or removing styrene monomer.

Polymerization Techniques

Premix Series. For radiation treatment of samples, two 3-in.-diameter by 6-in.-long test specimens were cast from each of six concrete–monomer mixes and a plain concrete control. After casting, one specimen of each pair was irradiated immediately at a radiation intensity of ~1.8×10^6 rads/hr for 24 hr to polymerize the monomers. The remaining seven specimens were removed from the molds at an age of 24 hr and then irradiated under similar conditions. On completion of the irradiation, the specimens were cured in water for 28 days. Two samples, in which alcohol was substituted for water, were cured in methyl alcohol.

For thermal-catalytic treatment of samples, experiments similar to those described above were conducted in which 5 wt.% benzoyl peroxide was added to the monomer and polymerization was initiated by heating to 176°F (80°C). Two 3-in.-diameter by 3-in.-long specimens were cast from each mix. One sample was immediately placed in an oven at 176°F (80°C) for polymerization. The second sample was polymerized after hardening for 18 hr. All specimens were then cured in water for 28 days.

Preformed Concrete–Polymer Series. For radiation treatment of samples, experiments are being carried out to measure the radiation requirements for the in-situ polymerization of monomers in preformed concrete. To date, a detailed study of methyl methacrylate has been completed, and data have been obtained for the bulk polymerization of several other monomers. Measurement of the radiation doses for these monomers in concrete will be contingent upon a favorable evaluation of the concrete–polymer properties now being measured.

The radiation-induced polymerization rate of methyl methacrylate in concrete has been measured as a function of radiation intensity. Concrete test pieces measuring 3 by 3 by 1 in., cut from test specimens supplied by the USBR, were used in all experiments. All pieces were dried to constant weight at 221°F (105°C) and impregnated with methyl methacrylate containing 15 ppm hydroquinone inhibitor with use of the "vacuum-soak" method described previously. The samples were then wrapped in polyethylene film and irradiated at a constant radiation intensity to a known total dose. A test-tube sample of the monomer was attached to the concrete for comparison of the rates. On completion of the irradiation, the sample was weighed to determine the amount of polymer and unreacted monomer in the specimen. The specimen was then crushed, evacuated to remove the unreacted monomer, and reweighed. The experiments were carried out as a function of radiation intensity and total dose. These data indicate higher polymerization rates in concrete than for the monomer alone. This is not totally unexpected, since it is known that in heterogeneous systems energy transfer effects at surfaces can take place in a radiation field and result in higher rates. At a radiation intensity of 3.4×10^5 rads/hr, complete conversion was obtained at a total dose of 1.8×10^6 rads. A total dose of 1.2×10^6 rads was required for complete conversion at a radiation intensity of 1.6×10^5 rads/hr.

A preliminary estimate of the radiation dose requirements for several other monomers has been obtained in survey-type experiments. The experiments were carried out for the purpose of producing test specimens for evaluation, and the materials probably received more radiation than was actually required. The data given in Table 4 should therefore be considered an upper limit.

Table 4 Radiation Requirements for Complete Polymerization of Monomers and Mixtures in Concrete

Monomer	Radiation intensity, rads/hr	Radiation dose, rads
Styrene	2.6×10^6	2.5×10^7
60% styrene–40% acrylonitrile	2.8×10^5	1.3×10^6
Methyl methacrylate + 10% trimethylolpropane trimethacrylate	2.5×10^5	1.0×10^6

Several series of concrete–polymer test specimens have been prepared for product evaluation by using thermal-catalytic methods. Monomers used were methyl methacrylate, styrene, and a methyl methacrylate–10% trimethylolpropane trimethacrylate mixture. Benzoyl peroxide concentrations of 5, 2, and 1 wt.%, respectively, were used. All concrete specimens were impregnated with the monomer–catalyst mixture by using the vacuum-soak method. After being dipped

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in a polymeric solution of the monomer under study and wrapped with polyethylene film to reduce vaporization loss, the samples were placed in an oven at 167°F (75°C). In all cases polymerization was completed within 2 hr, but the samples were left in the oven overnight for convenience.

A series of test specimens was prepared by the radiation-initiated polymerization of styrene containing 2 wt.% benzoyl peroxide. The catalyst was added to reduce the high radiation requirements for styrene. Survey experiments have indicated a maximum total dose requirement of 2.5×10^7 rads at an intensity of 2.6×10^6 rads/hr. Data from styrene test series indicate a maximum dose requirement of 2.08×10^7 rads at an intensity of 1.07×10^6 rads/hr. Assuming the polymerization rate to vary inversely as the square root of the intensity, the lower dose may be due solely to the reduced intensity. Additional data on this system are required.

Measurement of Properties of Concrete–Polymer Test Specimens

Types of Test

A comprehensive series of tests has been performed to evaluate concrete–polymer composites. The types of test performed were:

Compressive strength
Static modulus of elasticity
Tensile strength
Flexural strength
Flexural modulus of elasticity
Water absorption
Abrasion resistance
Cavitation resistance
Water permeability
Conductivity
Diffusivity
Thermal coefficient of expansion
Creep
Freeze–thaw durability
Resistance to sulfate attack
Resistance to acid attack
Resistance to distilled water at temperatures >100°C
Petrographic examination
Radiation stability

Test Procedures

In the full report, details of the procedures for performing tests for evaluating concrete–polymer composites are given. With each brief description of the procedure used, there is an accompanying discussion of the more important aspects of the test.

Preparation of Concrete–Polymer Test Specimens

As a means of systematically and thoroughly evaluating a number of liquid monomers, a detailed program for preparing and testing specimens was formulated. It consisted of preparing test quantities of 184 specimens for use with each monomer. To determine whether differences exist in the resultant properties, half of the samples were to be polymerized by radiation and the remainder by thermal-catalytic means. All samples are prepared and fog-cured for 28 days at the USBR, impregnated at BNL, and placed in test at the USBR at an age of 120 days. The specimens were divided into groups for initial screening (38 specimens), brine testing (45 specimens), and spares (9 specimens). To reduce the number of samples being exposed to the long-term brine tests, it was decided to determine which monomers would be tested in brine after evaluation of the results from the other screening tests.

Since preliminary testing had shown that the maximum loading could be obtained only with specimens that had been dried to constant weight, each of the screening tests was made with samples that had been oven dried at 221°F (105°C) to constant weight. These specimens are referred to in the text, tables, and figures as "dried" samples. An additional nine specimens that were stored in air prior to impregnation (referred to as "undried" specimens) were prepared for measurement of compression, tensile, and flexural strength, since it was thought that these properties would be affected the most by the degree of penetration and loading.

The impregnation procedure used in the survey series was modified for use in the test series experiments because of the increases in number and size of samples to be impregnated and the desire to reduce the volume of excess monomer being irradiated. Since preliminary experiments had shown that comparable polymer loadings and continuous surface coatings could be obtained, the following procedure was used to prepare all test series specimens.

1. Evacuate the dried or undried sample at a pressure of <3 in. Hg for 2 hr.

2. Soak in monomer for ~17 hr with a nitrogen blanket pressure of 5 psig.

3. Drain monomer.

4. Dip monomer-saturated concrete sample in a 10 to 15% polymer solution of the test monomer to coat the surface.

5. Wrap the specimen in polyethylene sheet to reduce the vaporization loss of monomer from the sample.

6. Polymerize to completion by radiation or thermal-catalytic means.

Results of Concrete–Polymer Tests

Complete summary listings of the properties of concrete–polymer materials, showing the differences with respect to controls and the differences between radiation and thermal-catalytic treatment, are given in Table 5 for methyl methacrylate monomer impregnant and in Table 6 for styrene monomer impregnant. The results are primarily for dried concrete with maximum polymer loading.

The remarkable improvements in properties compared with those of control specimens of untreated concrete may be briefly summarized as follows.

1. Compressive strength increases of 285% over control or a factor of almost four times the compressive strength of the control.
2. Tensile strength increases of 292%.
3. Modulus of elasticity increases of 80%.
4. Modulus of rupture increases of 256%.

Table 5 Concrete–Polymer Materials—Summary of Properties of Methyl Methacrylate-Impregnated Concrete (Dried Concrete Specimens Containing 4.6 to 6.7 wt.% PMMA)

	Control	Irradiation	Difference, %*	Thermal catalytic	Difference, %†	Difference, %‡
Compressive strength, psi	5267	20,255	285	18,161	244	−10
Modulus of elasticity, 10^6 psi	3.5	6.3	80	6.2	77	−2
Tensile strength, psi	416	1,627	292	1,508	262	−7
Modulus of rupture, psi	739	2,637	256	2,287	210	−13
Flexural modulus of elasticity, 10^6 psi	4.3	6.2	44	7.1	65	15
Water absorption, %	5.3§	0.29§	−95	3.58§	−33	1130
	6.4	1.08	−83	0.34	−95	−69
Abrasion, in.	0.0497	0.0163	−67	0.0147	−70	−10
Abrasion, g	14	4	−71	4	−71	0
Cavitation, in.	0.32	0.064	−80	0.020	−94	−69
Water permeability, 10^{-4} ft/year	6.2§	0§	−100	–	–	–
	5.3	0.8	−85	1.4	−73	75
Thermal conductivity at 73°F (23°C), Btu/(ft)(hr)(°F)	1.332	1.306	−2	1.265	−5	−3
Diffusivity at 73°F (23°C), sq ft/hr	0.0387	0.0409	6	0.0385	0	−6
Coefficient of expansion, in./(in.)(°F)	4.02×10^{-6}	5.36×10^{-6}	33	5.25×10^{-6}	31	−2
Creep for 800-psi load after 30 days, 10^{-6} in./in.	−95	+38	Neg. creep	+56	Neg. creep	–
Freeze–thaw durability, cycles, % weight loss	590; 26.5§	2,420; 0.5§	>310	–	–	–
	490; 25.0	750; 4.0	>53	750; 0.5	>53	−88
Hardness-impact ("L" hammer)	32.0	55.3	73	52.0	62	−6
Corrosion by sulfate solution, 300-day exposure, % expansion	0.144§	0§	−100	–	–	–
Acid corrosion in 15% HCl, 84-day exposure, % weight loss	10.4	3.64	−65	3.49	−66	−4
Corrosion by distilled H_2O 120-day exposure at 206°F (97°C)	Severe attack	No attack	−100	–	–	–

*Difference, % = $\frac{\text{radiation}-\text{control}}{\text{control}}(100)$.

†Difference, % = $\frac{\text{thermal}-\text{control}}{\text{control}}(100)$.

‡Difference, % = $\frac{\text{thermal}-\text{radiation}}{\text{radiation}}(100)$.

§From survey PLIC series; all other data from CP-1 test series. Results based on averages of 1 to 3 test measurements for each property.

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5. Flexural modulus of elasticity increases of 44%.
6. Freezing and thawing improved by >300%.
7. Hardness-impact ("L" hammer) increases of 73%.
8. Water permeability decreases to negligible values.
9. Water absorption decreases of as much as 95%.
10. Reduction of corrosion by distilled water and sulfate brines to negligible values compared with the severe attack observed on the control specimens.

The coefficients of expansion, specific heat, and diffusivity have been measured. Resistance to abrasion, cavitation, soluble sulfates, and acid corrosion has shown significant improvement.

Methyl methacrylate-impregnated concrete–polymer specimens subjected to creep tests exhibited an expansion under sustained load (negative creep). This is the opposite of the normal contraction in length (positive creep) of ordinary concrete. Further investigation of this highly unusual phenomenon of negative creep is necessary, since this property could have a significant influence on the design of concrete–polymer structural and prefabricated members.

Thermal-catalytically polymerized concrete–polymer had strength values 7 to 15% lower than ^{60}Co gamma-radiation polymerized concrete–polymer. Polystyrene impregnation of concrete gave ~30% lower strength values than polymethyl methacrylate. It was

Table 6 Concrete–Polymer Materials—Summary of Properties of Styrene-Impregnated Concrete (Dried Concrete Specimens Containing 4.2 to 6.0 wt.% PS)

	Control	Irradiation	Difference, %*	Thermal catalytic	Difference, %†	Difference, %‡
Compressive strength, psi	5267	14,710	180	9986	90	−32
Modulus of elasticity, 10^6 psi	3.5	7.7	120	7.4	110	−4
Tensile strength, psi	416	1,205	190	840	102	−30
Modulus of rupture, psi	739	2,388	223	1159	57	−51
Flexural modulus of elasticity, 10^6 psi	4.3	7.3	70	5.5	28	−25
Water absorption, %	5.3§	1.86§	−65	1.77§	−67	−5
	6.4	0.51	−92	0.70	−89	+37
Abrasion, in.	0.0497	0.040	−19	0.0365	−27	−9
Abrasion, g	14	9	−36	6	−57	−33
Cavitation, in.	0.320	0.035	−89	0.009	−97	−74
Water permeability, 10^{-4} ft/year	5.3	–	–	1.5	−72	–
Thermal conductivity at 73°F (23°C), Btu/(ft)(hr)(°F)	1.332	1.284	−4	1.305	−2	2
Diffusivity at 73°F (23°C), sq ft/hr	0.0387	0.0397	3	0.0408	5	3
Coefficient of expansion, in./(in.)(°F)	4.02×10^{-6}	5.09×10^{-6}	27	5.0×10^{-6}	24	−2
Creep for 800-psi load after 30 days, 10^{-6} in./in.	−95	+60	Neg. creep	+13	Neg. creep	
Freeze–thaw durability, cycles,	590; 26.5§	1810; 33§	207	–	–	–
% weight loss	490; 25.0	620; 6.5	>27	620; 0.5	>27	−92
Hardness-impact ("L" hammer)	32.0	48.2	51	50.1	56	4
Corrosion by sulfate solution, 300-day exposure, % expansion	0.144§	0§	−100	–	–	–
Acid corrosion in 15% HCl, 84-day exposure, % weight loss	10.4	5.5	−47	4.2	−60	−24
Corrosion by distilled H_2O, 120-day exposure at 206°F (97°C)	Severe attack§	No attack§	−100	–	–	–

*Difference, % = $\frac{\text{radiation} - \text{control}}{\text{control}}(100)$.

†Difference, % = $\frac{\text{thermal} - \text{control}}{\text{control}}(100)$.

‡Difference, % = $\frac{\text{thermal} - \text{radiation}}{\text{radiation}}(100)$.

§From survey PLIC series; all other data from CP-2 series. Results based on average of 1 to 3 test measurements for each property.

also noted that cracking of the concrete occurred in undried specimens impregnated with styrene and polymerized by radiation. There was no cracking with styrene impregnation of dried and irradiated concrete or with styrene impregnation of dried and undried thermally treated concrete.

Radiation and thermal treatment of unimpregnated concrete gave no improvement in structural properties. The improvement in properties is thus attributed to polymer impregnation.

A series of test specimens with cross-linked polymethyl methacrylate has been prepared. The test results are not yet available.

In the case of premixed concrete–polymer preparations, improvements in compressive strength and water absorption were obtained with styrene and acrylonitrile monomer additions. The maximum improvement (87% in compression with acrylonitrile) was much lower than that with impregnated preformed concrete. In the case of methyl methacrylate premixed, there was a marked decrease in compressive strength. Substitution of methyl alcohol for water inhibited the hardening of concrete.

High-strength OSW concrete (type V cement, air-entrained with fly ash and aggregate) impregnated with methyl methacrylate gave the highest value of compressive strength, 22,630 psi, equivalent to a 164% improvement over untreated OSW concrete.

Fundamental Studies

The fine structure of the concrete–polymer material is being examined by optical and electron microscopy and petrographic techniques. Fundamental studies are in progress to obtain an understanding of the bonding of the polymer to the concrete on a microscale. These studies show that for preformed concrete at higher loadings the impregnation was fairly complete and the polymer coated most of the internal surfaces of the void passages in the concrete. In the premix concrete the polymer distribution varied from poor to good.

Comparison of Concrete–Polymer with Other Materials

It is interesting and instructive to compare the strength properties of the concrete–polymer material with those of concrete, polymer, and other construction materials and to obtain some rough cost comparisons.

On the basis of tensile strength, aluminum shows up best on a strength to weight ratio, and steel is best on a cost to strength ratio. Concrete–polymer on a cost basis is better than either concrete alone or polymer alone. In the case of compressive strength, polymer alone is exceeded only by glass on the basis of strength to weight ratio. On a cost to strength ratio, concrete–polymer is far superior to polymer alone and to concrete alone; it is exceeded only by glass. Although the compressive strength of glass is extremely high, it is very brittle and strongly dependent on its surface condition, which is susceptible to abrasion and is therefore highly unreliable. In terms of the ratio of modulus of elasticity to weight, steel and aluminum are better than concrete–polymer. However, from a cost point of view, concrete–polymer and concrete are far superior to the others.

In terms of tensile and compressive strength, the cost to strength ratio for concrete–polymer is about half that of finished concrete. This is for a 6% loading of 20 cents/lb methyl methacrylate. Assuming the radiation processing cost to remain approximately constant, more expensive monomers could be considered for impregnation, up to the break-even cost for finished concrete alone. The break-even cost for monomer under the above conditions is thus about $1.20/lb. These cost data indicate that the more expensive monomers need not be eliminated from consideration.

When tensile strength is required, concrete is usually reinforced with steel, which, as the cost to strength ratio indicates, is much more economical for this purpose than any of the other materials, including concrete–polymer. However, concrete–polymer should be most useful when compressive force is the dominant structural factor.

Advantages other than strength properties should make concrete–polymer still more competitive. Improvements in durability properties such as reduced water absorption, resistance to freeze–thaw, corrosion resistance, and aesthetic value must be included as factors in assessing the total value of the material for construction purposes.

It should be noted that the above is only a preliminary attempt, with the data on hand, to evaluate concrete–polymer versus other more common construction materials from a fundamental point of view. The general conclusion is that the use of concrete–polymer for structural purposes, under certain conditions, appears economically reasonable, and the material is therefore well worth further develop-

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ment. To choose a material for a specific application, much more definitive test and cost information would be required.

Applications Development

With some of the techniques of preparation of concrete–polymer material developed and some measurements of the properties of the composite material now available, investigation of specific process and product applications can be initiated.

Concrete Pipe

Concrete pipe is used in the transportation of irrigation water, sewage, and municipal and industrial water. Total annual sales run ~$500 million for a production of about 200 million lineal feet of concrete pipe per year. The possible advantages of concrete–polymer are due either to the improved structural properties for purposes of saving on material costs and increased pressure rating of pipe, or to improved durability, especially against water corrosion. The concrete–polymer pipe should be compared with reinforced- and prestressed-concrete pipe and under corrosive conditions with glazed tile, Duriron, and plastic composition pipe. A preliminary conceptual design of a concrete–polymer pipe plant and cost estimate[2] indicates some potential. A more detailed cost analysis is being prepared.

Housing Applications

Concrete–polymer material may have some application in the construction of low-cost housing. Concrete is extensively used as a low-cost material in the building industry. By increasing its strength and decreasing its water absorption, marked reductions in material cost and much improved durability may be achieved. Supporting or load-bearing columns could be made smaller or carry additional building height. Prefabricated panels could be made lighter and stronger. Lightweight aggregate concrete wall panels for prefabricated structures could serve both as wind breaks and as thermal insulation. In addition, dyes can be used to provide an aesthetically pleasing permanent finish requiring little maintenance.

It has also been suggested that concrete–polymer be used in livestock and produce processing plants for walls or floors to provide sanitary nonabsorbent surfaces with low maintenance costs.

Desalination Plants

The Office of Saline Water together with the U. S. Bureau of Reclamation is investigating the use of concrete as a construction material in seawater-desalination plants.[1] At 290°F (143°C) and above, concrete corrodes in brine. Distilled water is also corrosive; it dissolves the concrete matrix. Many types of protective coatings have been tried, but all have proved unsatisfactory, either because they were not corrosion resistant or because the surface coatings did not bond adequately to the surface of the concrete and the coating became separated and peeled off. An impregnation of polymer deep into the surface of the concrete to form a concrete–polymer coating could possibly provide adequate corrosion resistance with the necessary strong bonding between the protective polymer material and the concrete base. Cross-linking and thermosetting monomers are indicated for the high-temperature conditions obtained in desalination plants. For this application physical and corrosion tests will have to be made at elevated temperatures.

Underwater Applications

Increasing effort is being expended in the development of ocean technology for both commercial and scientific purposes. The Naval Civil Engineering Laboratory[11] is testing concrete as a construction material for such applications as pressure hulls for underwater and ocean installations. The limitations of concrete are its compressive strength and water permeability. Concrete–polymer material is well suited to this application, since the depth to which vessels can go is directly related to the compressive strength, and the permeability is reduced to negligible values. As an example, it is calculated that a high-strength concrete sphere 5½ ft in outside diameter with a 4⅛-in.-thick wall is suitable for external pressures down to a depth of 1000 ft. The use of concrete–polymer material allows a sphere of the same dimensions to reach a depth of 2300 ft. In addition to its impermeability, the nonconductive nature of concrete–polymer allows transmission of electromagnetic signals for communication purposes. The resistance of concrete–polymer to seawater corrosion should be very good. Surface buoys constructed of concrete–polymer would also be less expensive than steel or aluminum buoys. Concrete–polymer should also be suitable for underwater habitats in off-shore installations. As the new field of ocean technology grows, the application of concrete–polymer may become important.

Prestressed-Concrete Pressure Vessels

Much effort is going into investigations on the use of concrete in large nuclear reactor power plants.[12] The size of nuclear plants is increasing because the larger sizes offer more favorable economic conditions. Vessels constructed of concrete have the advantage over steel in that very large reactor vessels can be field-constructed with no size limitations and improved plant safety features can be incorporated, especially in gas-cooled systems. The large concrete vessels are steel reinforced and prestressed. The possibility arose of using concrete–polymer for improved strength, with a consequent saving in materials. Preliminary calculations indicated that not much improvement can be obtained for the inner section of the vessel, where the concrete is mainly under compression. However, a limitation on the concrete exists on the outer section of the vessel, where the concrete is mainly under tension. Another limitation is due to the many steel bearing pads required to tie down the steel tendons. The size and number are limited by the compressive strength of the concrete. The use of high-bearing-strength concrete–polymer on the outer few inches of the concrete vessel would permit a marked reduction in the area and weight of the anchor plates. Considering the physical spacing of these bearing pads and the sizable investment they represent, the use of concrete–polymer in the outer section of the reactor would be a substantial improvement. The reactor vessel would in effect be case-hardened. Additional advantages would be decreased gas and water permeability and generally less deterioration of the outer surface. The radiation levels in the outer section are very low and, in view of the data obtained on the radiation stability of concrete–polymer, little deterioration in properties is to be expected. Consideration has yet to be given to field impregnation and polymerization techniques.

Antifragmentation Concrete

Several tests on the effects of high-velocity projectiles on plastic-impregnated unreinforced-concrete blocks have indicated a marked improvement in the impact resistance and the antifragmentation properties of concrete. A 30-caliber bullet fired from a distance of 50 yd at a 13.5- by 7.5- by 3.5-in. concrete–polymer block was stopped after penetrating 2 in. within the block, with little fragmentation on the reverse side. An unimpregnated concrete control specimen was pierced and showed severe fragmentation. A number of applications as armor material are suggested by this test. Much additional testing of this antifragmentation property of concrete–polymer would be required to determine its true value.

Aesthetic Applications

A red aniline dye dissolved in the liquid monomer was used to determine monomer penetration. A polished cross section of the dyed concrete–polymer material had a lustrous red surface. A variety of colored dyes could be used to give the concrete–polymer material a "terrazzo" effect for use in flooring, wall facings, or works of art. Since the dye is incorporated throughout the impregnated concrete, maintaining the appearance of the surface should be an easy matter. The surfaces of art objects could probably be preserved for longer periods of time.

Because of the striking improvement in structural properties and durability, many more applications of concrete–polymer are possible. The combined ingenuity of the materials producer and the structural engineer should result in valuable practical product applications of concrete–polymer.

Recommendations

The following recommendations are made for the future program.

1. Continued effort should be expended on developing and optimizing concrete–polymer preparation techniques. These include drying, impregnation, coating, and initiation of polymerization by both radiation and thermal-catalytic means.

2. Additional monomers should be subjected to concrete–polymer preparation and investigated in survey and thorough test experiments. These should include vinyl monomers, polyesters, epoxies, and cross-linking agents in vinyl monomers.

3. Different types of concrete should be investigated, including nonair-entrained concrete, high-density concrete, lightweight aggregate concrete, steel- and fiber-reinforced concrete, and cement mortar alone. Concrete of varied water, cement, sand, and aggregate compositions should be impregnated and tested. The curing times of the concrete used in producing the composite should also be investigated.

4. Premixed concrete preparations in which the monomer is added to the freshly mixed concrete and either radiation or thermally treated should be further evaluated.

5. The strength properties of concrete–polymer composite should be measured at higher temperatures.

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6. Plant design and evaluation studies should be made on applications of potential interest and economic value.

7. Fundamental microstructure studies should be continued to aid in the understanding of the properties of concrete–polymer composites. (R. E. Greene)

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*Radiation Vulcanization of Natural Rubber in the Latex Phase: Study of a 1-Tonne/hr Pilot Plant**

The reasons are given for selecting an accelerator for the basic studies of a radiation-vulcanization pilot plant. Also considered were a ^{60}Co source, a nuclear reactor, and spent nuclear fuel in the cooling stage. The ^{60}Co source was rejected because of the large quantity—2×10^6 curies—required. Equipment tests that were made are described, and properties of irradiated rubber are recorded. (MG)

*In French, by P. Leveque, J.-R. Puig, and H. Roudeix, Saclay Report CEA-R-3345 (1968) (76 pp.).

Section

V

Isotopes and Radiation Technology

Isotopic Heat and Power Development

A Summary of Recent Work by the French in the Development of Isotopic Thermoelectric Generators

By D. N. Hess

Abstract: *The current status of French isotopic thermoelectric generators—Geter-2B, Marguerite II, and Marguerite 20—is reviewed.*

Design considerations, materials specifications, operating parameters, on-site installation, and performance are among the subjects discussed. Power demands for operational and projected generators for specific applications are listed. The fabrication of the $^{90}SrTiO_3$ heat sources for a number of different generators is summarized, with special emphasis on the materials, their requirements and evaluation, and on the preparative procedures involved.

In several recent issues of *Isotopes and Radiation Technology*[1-4] a number of developments in European isotopic thermoelectric generators were reported. This article is a brief review of several aspects of the French activities in this area, as published in *Isotopic Generator Information Centre Newsletter*, No. 7, November 1968 (CEA–ENEA).*

Generator Applications and Power Requirements

In a summary by the Société Nationale d'Étude et de Construction de Moteurs d'Aviation (SNECMA) entitled "Applications of Isotopic Generators," those generators already in worldwide service are reviewed with respect to their applications for submarine cables, radio links, beacon lights, acoustic underwater warning devices, radio beacons and navigational aids, responder buoys, oceanographic research, weather stations, and offshore oil fields. From information supplied by the Compagnie Française des Pétroles, SNECMA gives examples of offshore oil-field equipment that could be powered by isotopic generators. In regard to radio and acoustic communication, they discuss the power requirements of transmitter–receivers operating under intermittent conditions and the dependence of power requirements on the number of interrogations and orders, the distance involved, and the acceptable quality of transmission. Estimates of these requirements are listed below.

Conditions	Power requirements
Radio communication	
Single well; 864 interrogations, 10 remote-control orders	2.18 watts
Cyclical well, platform type; ½-time transmitter, ½-time receiver	5.16 watts
Platform-to-ground relay transmitter–receiver	5 to 60 watts
Acoustic communication (sonic interrogation)	
Sonic transmission, 15 min daily	5 watts
Master transmitter–receiver, 1 hr daily	21 watts

Power requirements were also listed for such functions as:

*Authors and titles of papers from which the information for this report was obtained are as follows:

R. Brouée, *Geter-2B. Thermoelectric Isotopic Generator.*

Société Nationale d'Étude et de Construction de Moteurs d'Aviation and Compagnie Française des Pétroles, *Applications of Isotopic Generators.*

P. Thery, *Marine and Space Generators Developed by SNECMA.*

J. Freund, *"Marguerite II" Generator.*

J. Fradin, *The Fabrication of Strontium-90 Titanate Radioactive Sources for Isotopic Generators.*

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Conditions	Power requirements
Platform-buoy beacon lights	
3-mile range; 20% on-time, 12 hr daily	1 watt
5-mile flashing light	4 watts
8-mile flashing light	20 watts
Fog horns	
180-watt horn; 5-sec on-time, 15-sec interval	45 watts
Underwater ultrasonic warning devices	
3-mile range	1 watt
Radio beacons for helicopters	
Remote controlled, functioning on approach of helicopter; 1 hr daily	1.5 watt
Valves	
Electrical actuation; 2 3/8-in. I.F.P./F.S. valve; 48 times daily	20 watts
Hydraulic actuation; 4-in. W.K.M. valve;	
Once per 24 hr	0.42 watts
24 times daily	10 watts
(Remote control; additional requirements)	2 watts
Hydraulic actuation; type F-Cameron valve	2 x W.K.M.
Obturators	
18-in. G.H. Hydrid; once daily	40 watts
20-in. Cameron ram type; once daily	15 watts

Geter-2B*

The Geter-2B generator, a second-generation version of Geter-1,[1] is fueled with $^{90}SrTiO_3$ and is being built by S.E.M. Hispano-Suiza under a Euratom contract. Table 1 shows the specifications of the device, and Fig. 1 shows the completed machine. Geter-2B generates ~300 mw(e) from 2500 curies of ^{90}Sr.

Marguerite Series

The Marguerite II and Marguerite 20 generators, developed by SNECMA, are intended for long-term operation as surface (light) and underwater (ultrasonic) buoys. The strontium titanate heat sources are encapsulated in double-walled leaktight cans fabricated of Hastelloy alloy C for Marguerite II and Haynes alloy No. 25 for Marguerite 20 which are resistant to seawater corrosion, experience no oxidation at high temperatures, will not undergo brittle fracture, have good welding properties, and have mechanical properties that are satisfactorily retained at operating temperatures. The double-walled containment vessel is capable of withstanding pressures of 1000 bars. Main design considerations were concerned with the hot-cell loading of the radioactive source, radiological protection to civilian handlers, minimization of heat losses to the marine environment, and ensured safety during transportation and operating lifetime.

Marguerite II

Some pertinent data concerning the Marguerite II generator are shown in Table 2.

Table 1 Specifications of the Geter-2B Generator

Heat source	
Material	$^{90}SrTiO_3$
Activity	171 curies/watt(t)
Specific mass	3.6 g/cm^3
Specific power	0.64 watt/cm^3
Thermal conductivity	4.85 watts/(m)(°C)
Electrical power	420 mw
Thermoelectric modules	
Type	Bi_2Te_3
Dimension of *n* and *p* thermoelements	Diameter, 3 mm, length, 8 mm
Number of thermocouples	12
Hot-junction temperature	220°C
Cold-junction temperature	30°C
Electrical properties before conversion stage	
Open-circuit voltage	864 mv
Internal resistance	427 milliohms
Voltage under load	500 mv
Nominal power	426 mw
Maximum power (with appropriate load)	436 mw
Nominal current rating	850 ma
Electrical properties after conversion stage	
Converter efficiency	70%
Voltage under load	6 volts
Nominal current rating	47 ma
Nominal power	295 mw
Other characteristics	
Thermal power required	14.8 watts(t)
Efficiency before conversion	2.8%
Efficiency after conversion	2%
Mass of strontium titanate	83 g
Activity of strontium titanate	2500 curies

* ". . . there exists only one Geter, the Geter-1B. However, some of these devices are called Geter-2B." Private communication from Gustav Rosner, CEA–ENEA, who kindly reviewed this report and to whom the author wishes to express his appreciation.

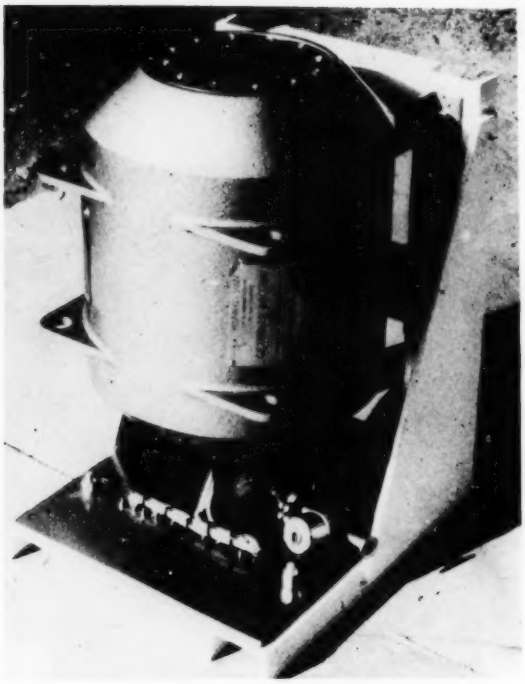

Fig. 1 Geter on its support, viewed from the generator side.

Table 2 Design Characteristics of Marguerite II

Vessel	
Diameter	0.350 m
Height	0.450 m
Mass of generator	450 kg
External shell	Stainless steel
Corrosion, seawater	4×10^{-2} mm/year
Pellet	
Radioactive material	$^{90}SrTiO_3$
Dimensions	25 mm diameter by 12 mm high
Power	650 curies (4.4 watts)
Solubility, seawater	3.2×10^{-6} g/(cm^2)(day)
Source container	
Wall thickness, inner can	1 mm
Wall thickness, outer can	0.05 mm
Clearance	
Between source and inner can	0.05 mm
Between inner and outer cans	0.05 mm
Atmosphere	Argon
Material	Hastelloy alloy C
Corrosion, seawater	2.5×10^{-3} mm/year
Heat conversion system	
Thermoelectric material	Bi_2Te_3
Junction temperatures (on land)	205°C, 25°C
Thermopile power under load	125 mw @ 0.5 volt
Efficiency before conversion	2.8%
Converter	d-c/d-c
Converter output	106 mw @ 5.4 volts
Efficiency	84%
Expected service life	>5 years
Radiological safety	
Radiation shielding	Lead and steel
Dose rate	
1 m from source	0.4 mrad/hr
At side wall of generator	12 mrads/hr

Thermal Conversion System. A 2-mm-thick molybdenum (chosen because of its good creep properties and thermal conductivity) heat collector conducts the heat to the connecting straps of the thermopile. An automatic clock device actuates this heat collector when the core-loading tools are withdrawn from the assembly. Heat from the radioactive source is converted to electricity by 18 *n*- and *p*-type bars of solid solutions of doped bismuth telluride. Copper strap brazed onto the thermoelectric material provides electrical connection between the end of a *p*-bar and the end of an *n*-bar. Assembled side by side, the bars are electrically insulated by a heat and gamma-radiation-resistant epoxy resin. The thermopile is rectangular in shape, measuring 26 by 17 by 17 mm, and is held in place between the collector and a heat sink. The latter is a double-walled steel container that has been filled with lead; it is enclosed within the cavity provided by the stainless-steel-clad biological shield and conducts unconverted heat to the outer shell and into the seawater environment after immersion. The radiological protection afforded by the shield results in a dose rate of <0.4 mrad/hr at a distance of 1 m, which is the accepted norm for civilian workers. The thermopile is electrically insulated from the collector and the heat sink by an alumina packing to avoid short circuits. Since the thermoelectric material may be oxidized at high temperature, the generator is maintained in an inert atmosphere.

A thermal insulation block, which maintains the heat source at a temperature level that affords optimum conversion efficiency while protecting the thermoelectric elements from any possible diffusion of the dopants, consists of:

1. A spacer of asbestos-based structural insulating material that holds the heat sink and thermopile in position under the severest operating condition that can be anticipated.

2. Microporous insulation washers—in a neutral atmosphere—which fit into any interstices between

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the thermopile heat sink and the spacer and which are compacted by lateral webs and a front plate.

3. A microporous plug to stopper the hole through which the source is loaded.

A sliding hatch is the final closure on the generator core. Electrical outputs are obtained from watertight connectors attached to the heat sink.

The generator and its transport container were constructed to withstand fierce fires, 10-*g* impacts, vibrations up to 50 cps under a force of 3 *g*, and immersion to a depth of 1000 m. The generator (Figs. 2 and 3) was loaded Jan. 25, 1968, by the Commissariat a l'Énergie Atomique (CEA) and placed in operation on Feb. 21, 1968, under the responsibility of the Compagnie Française des Pétroles (CFP). It lies under 80 m of water and supplies, at the surface, a flashing light that has a nighttime visibility of 5 km, and its underwater ultrasonic generator has a detectability of 1.5 km. Its continuous operation of 5000 hr has fully confirmed the feasibility of reliable autonomous power units for oceanographic use and for exploitation of the resources of the sea. Figures 4 and 5 show the generator being immersed and its position on site, respectively.

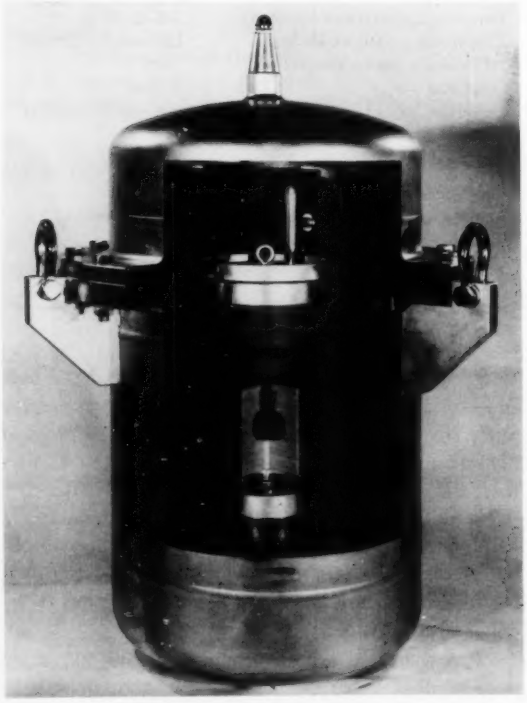

Fig. 2 Cutaway model of Marguerite II showing the general arrangement of the generator. The large amount of biological shielding around the core may be seen.

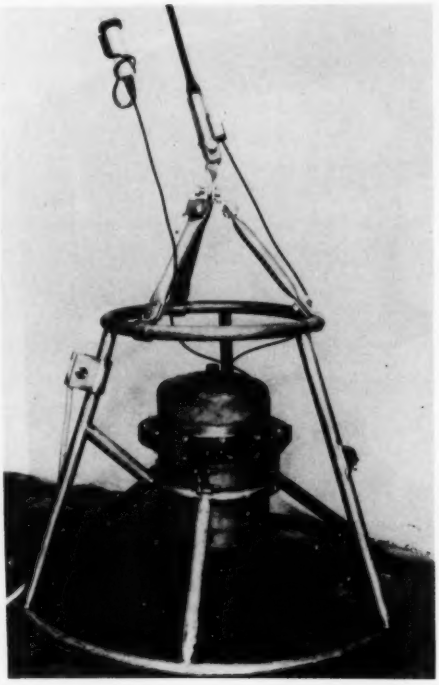

Fig. 3 Marguerite II generator on its immersion cradle. One of the two electrical outputs supplies an ultrasonic generator (to be seen in the top left-hand corner of the figure) and the other a flashing light on a floating buoy.

Marguerite 20

Design and development of an improved version of Marguerite II are in progress. The Atomic Energy Division of SNECMA, the Physico-chemical Research Department of Cie. Générale de Télégraphie sans Fil (CSF) and the Radioisotopes Department of the Nuclear Energy Research Center (Saclay) are intimately involved in the program. In order to provide 20 to 35 watts, depending on the user's requirements, the radioactive source will be in excess of 10^5 curies, the source temperature will be 1000°C, and Ge–Si thermoelectric couples will be used as converters. Accordingly, these factors have required certain design changes. The generator is intended to have a service life of 5 years and to be immersed at 1000 m. As with Marguerite II the objectives are simplicity in assembly, loading, and use; high reliability; and complete operational safety.

Fig. 4 Marguerite II being immersed for the first time, on Feb. 21, 1968.

Uses envisioned for this higher powered device are in applications involving servo systems, telemetry, and transmission. It will be placed in service by the CFP at an underwater wellhead.

Extraterrestrial Generator

The Atomic Energy Division of SNECMA and the Physico-chemical Research Department of CSF, under contract to the Centre National d'Études Spatiales, are jointly designing and developing a prototype isotopic generator for use in space vehicles. In an experimental chamber, simulating a space environment, the generator was tested, using electric power to simulate the 5×10^4-curie strontium titanate source. The complete unit is 320 mm high, 185 mm in diameter, and weighs 12.5 kg. Its 32 Ge–Si converters are assembled radially between the outer wall of the radioactive-source container of heat-resisting alloy and the outer radiator. The power developed exceeded 12 watts, corresponding to an efficiency of 3% and a specific mass of 1 kg/watt. It has been in continuous, completely stable operation for more than 4000 hr at a temperature in excess of 900°C. Figure 6 shows the generator during assembly; and the completed generator, just prior to testing, is shown in Fig. 7.

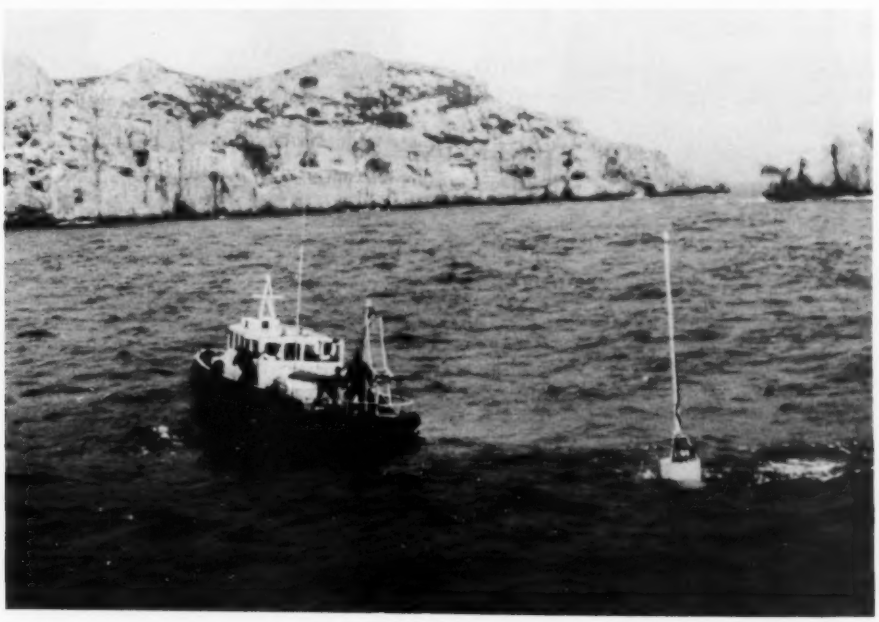

Fig. 5 Buoy and light supplied by the Marguerite II generator.

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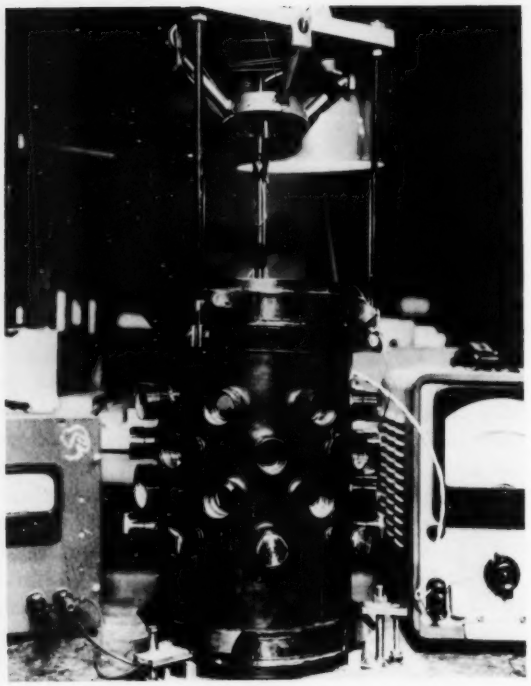

Fig. 6 Space generator during assembly.

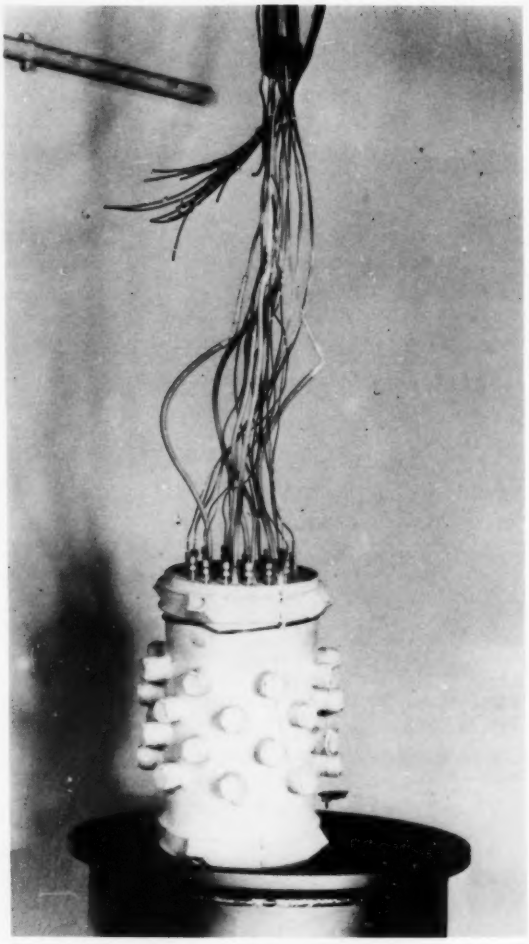

Fig. 7 Assembled generator being placed in the experimental chamber.

Radioactive Heat Source

The fabrication and encapsulation of the $^{90}SrTiO_3$ sources for the thermoelectric generators have involved extensive fundamental and developmental studies. Table 3 lists the pertinent physical, corrosion, and power characteristics of this stable ceramic refractory.

Design Considerations. Thirteen different measurements that were performed on the substance are indicated in Table 4, although not all tests were exhaustive.

In Table 5 are listed some of the source-containment criteria that had to be examined. The conditions imposed pertained particularly to thermoelectric generators that reach temperatures as high as 1100°C (in contrast to thermodynamic devices that operate in the region of 150 to 700°C).

Sixteen alloys were evaluated, all of which had disadvantages (Table 6).

Some of the problems inherent in each are as follows:

1. Monel and Inconel corrode under the contemplated service conditions.

2. Niobium and tungsten are oxidized after 1000 hr at 1000°C.

3. Hastelloy and Monel lack high-temperature stability.

4. Molybdenum, tungsten, and others acquire brittleness upon welding.

5. TD Nickel exhibits microporosity, undergoes a phase change at 800°C, and apparently would require forging, precision casting, or chemical machining to achieve finished dimensions.

6. FS-85 and T-111 are prohibitive in cost, and most others are quite expensive.

7. All are very difficult to work, particularly to machine and to weld.

Table 3 Significant Properties of Strontium Metatitanate Radioactive Heat-Source Ceramic

Melting point	1900°C
Sintering point	1350°C
Density	~5 g/cm^3
Solubility in water	3.2×10^{-6} g/(cm^2)(day)
Thermal conductivity between 800 and 1000°C	10^{-2} cal/(cm)(sec)(°C)
Specific power	0.23 watt/g
Specific activity	32 curies/g
Power density	6.7×10^{-3} watt/curie
Power per unit volume	0.8 watt/cm^3

Table 4 Physical Parameters Investigated on $^{90}SrTiO_3$ Ceramic

Thermogravimetry
Powder compression
Temperature of sintering
Density
Microhardness
Porosity
Thermal conductivity
Electrical conductivity
Expansion characteristics
Young's modulus as a function of temperature
Compressive strength as a function of temperature
Resistance to thermal shocks
Dimensional deformation—shrinkage after sintering

Table 5 Requirements of Alloy for Containment of Radioactive $^{90}SrTiO_3$ Heat Source

Satisfactory mechanical properties at high temperatures
Absence of brittleness after welding
Absence of brittleness caused by radiation
Sufficient resistance to gaseous corrosion at high temperatures and in various media
Sufficient resistance to seawater corrosion
Good thermal conductivity
Absence of diffusion of the encapsulation elements into the active compound, and vice versa, at high temperatures

Table 6 A List of Potential Containment-Vessel Alloys Tested

Molybdenum	Tantalum	Hastelloy X	TD Nickel
Tungsten	Monel K 500	Hastelloy F	FS-85
Niobium	Inconel 600	Haynes 25	T-111
Platinum	Hastelloy C	Haynes 152	TZM

Variations in service conditions and intended uses for a generator necessitate a thorough investigation of the encapsulation material—for example, whether lead telluride or germanium–silicon thermoelectric elements are being considered, or whether the generator is to be used for marine applications (water cooled), or for terrestrial or space applications (liquid-metal cooled). Corrosion could be a dominant consideration, depending on these differing situations. Moreover it is essential that the compatibility of the different materials be thoroughly examined with respect to diffusion, differential expansion, and placement of components.

Fabrication. The cells and equipment, such as grinders, sintering furnaces, can cutters, and automatic welders at the CEA site for the preparation of the ceramic pellets are described. (Since these facilities are more or less standard at all atomic installations, they will not be discussed in detail.) In Figs. 8 to 10, some of the more complex devices are shown.

It was of interest to note that a helium spectrometer was used to check the integrity of the cans, that total residual surface contamination was limited to 500 μc, and that a final check of the power of the source was made by using a calorimeter.

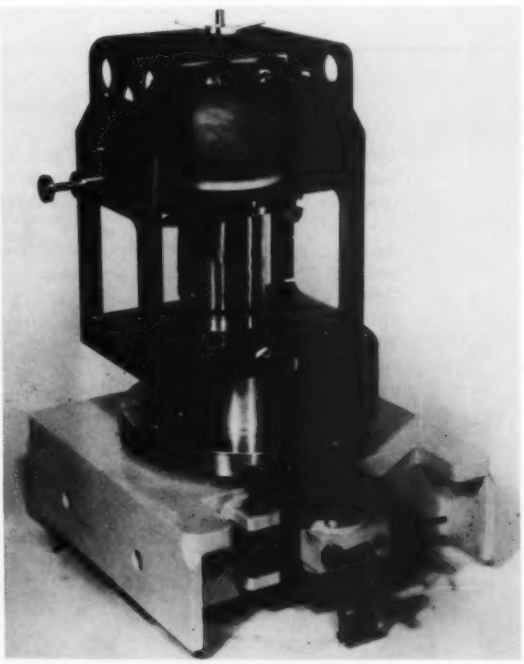

Fig. 8 Strontium titanate powder grinder.

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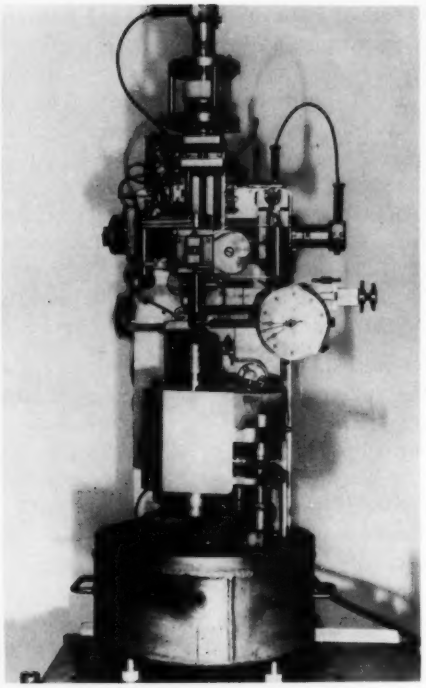

Fig. 9 Remote-controlled drilling machine.

Sources produced to date are:

750 curies for Marguerite II	(SNECMA–CFS)
2,500 curies for Geter-1B	(Hispano-Suiza)
14,000 curies for Gisete 4	(Alcatel)

By late 1968 two higher power sources were to have been fabricated:

125,000 curies for Marguerite 20	(SNECMA–CSF)
85,000 curies for Gisete 5	(Alcatel)

The former was also to have been loaded by late 1968; loading of the latter was expected to be completed in early 1969.

Future Plans

Finally, it is reported that work is in progress on thermodynamic generators utilizing motors or turbines (Rankine or Stirling cycles) to supply energy of the order 1 kw.

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1. Jerome G. Morse, Isotopic Power in Europe-II, *Isotop. Radiat. Technol.*, 6(1): 100-108 (Fall 1968).
2. F. E. McKinney, English Colloquium on Isotopic Thermoelectric Generators, *Isotop. Radiat. Technol.*, 6(2): 188-197 (Winter 1968–1969). [Summary of *Isotopic Generator Information Centre Newsletter*, No. 6, May 1968, (CEA–ENEA)].

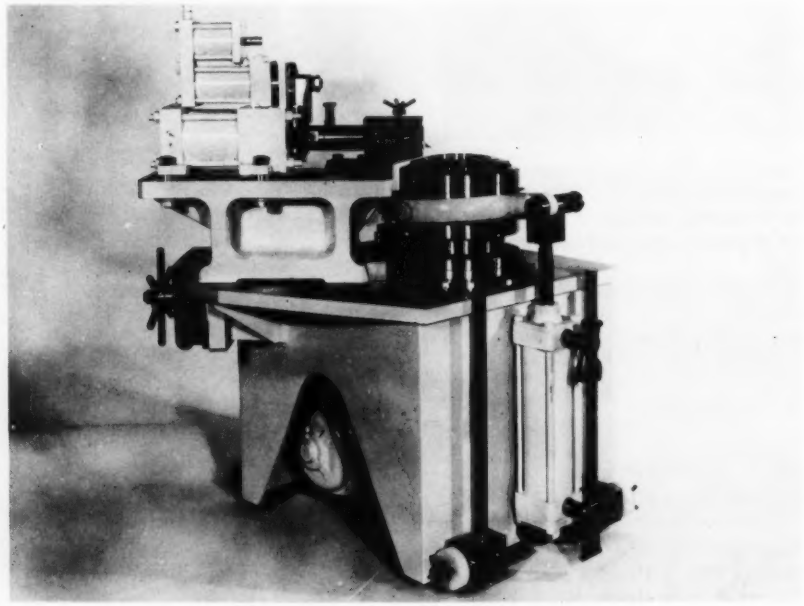

Fig. 10 Machine for cutting source capsules.

3. G. Rosner, Isotopic Generators in the Federal Republic of Germany, *Isotop. Radiat. Technol.*, 6(2): 197-199 (Winter 1968–1969). [*Isotopic Generator Information Centre Newsletter*, No. 5, March 1968 (CEA–ENEA).]

4. Martha Gerrard, First ^{242}Cm Isotope Battery in Europe, *Isotop. Radiat. Technol.*, 6(3): 321-322 (Spring 1969). [From *Euratom*, 7(3): inside back cover (September 1968).]

Mutations in Plant Breeding*

The second FAO/IAEA meeting on coordination of research on the use of neutrons in seed irradiation was held in Vienna in December 1967. The report of the meeting† includes the 19 papers that were presented together with new recommendations of the panel, based on knowledge acquired since the first meeting.

An extensive review of chemical- and radiation-induced mutations in plant breeding was presented at a 1964 meeting in Rome,‡ attended by 158 people from 32 countries. Convincing evidence was presented on the value of artificial mutations, both chemical- and radiation-induced, in plant breeding. (MG)

*Earlier publications reviewed in *Isotopes and Radiation Technology*, 5(4): 313-314 and 6(3): 319.

†*Neutron Irradiation of Seeds, II*, Technical Reports Series, No. 92, Report of a Panel Meeting, International Atomic Energy Agency, Vienna (STI/DOC/10/92).

‡*The Use of Induced Mutations in Plant Breeding*, report of the FAO/IAEA Technical Meeting, Rome, 1964, Pergamon Press, Inc., New York, 1965 (832 + ix pp.; $45.00; 66 papers and discussions).

Transportable Beta Irradiator for Medical Use

A transportable beta irradiator has been developed which is useful for extracorporeal irradiation of blood and lymph in leukemia therapy and in suppression of immune antibody response to organ transplants and grafts. The equipment is available from Radiation Machinery Corp., P. O. Box 177, Parsippany, N. J. 07054.

The irradiator is 23 in. long by $16\frac{3}{4}$ in. wide by $10\frac{1}{4}$ in. high, and it weighs 360 lb. It is small enough to be mounted on a table or cabinet fitted with wheels and pushed about by an operator. The radiation source is 20 curies of ^{90}Sr–^{90}Y ($T_{\frac{1}{2}}$ = 38 years; 2.27-Mev beta). Blood or lymph is shunted, via a tube, from the body circulation through the irradiator. Fluid flowing through a 2.6-mm-wall tube at 100 ml/min receives a radiation dose of up to 250 rads per transit. At a flow rate of 6 ml/min—the rate of lymph flow—a dose of 4300 rads per transit is delivered. The 20-curie ^{90}Sr source delivers more radiation to the blood than a 2000-curie ^{137}Cs or ^{60}Co gamma source (based on the amount of energy absorbed in 2.6 mm of unit-density material), which, with necessary shielding, would weigh several tons. The external radiation dose from the ^{90}Sr irradiator is less than 2 mr/hr. (MG)

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Section

VI

Isotopes and Radiation Technology

Isotopes Around the World

Sweden's Radioisotope-Powered Navigational Light

Sweden's first radioisotope-powered navigational light,[1] installed in 1967, is located on submerged rocks in the Stockholm archipelago. The power source, a RIPPLE-VI generator[2-4] from Britain, is fueled with 8 thousand curies of $^{90}SrTiO_3$ and is expected to operate for 5 to 10 years without refueling.

The fuel is in the form of two encapsulated cylinders, each 3 cm in diameter and 4 cm long, placed end to end (Fig. 1). The free ends of the cylinders contact bismuth telluride thermal elements for converting the radioisotope-decay heat to electricity. The fuel unit is surrounded by thermal-insulation material, which, in turn, is surrounded by a radiation shield of depleted uranium. Maximum radiation levels are 150 mr/hr at the surface of the source and 5 mr/hr at 100 cm from the center.

The unit is installed in a conventional navigational-light structure (Fig. 2), a photograph of which was shown in an earlier issue of *Isotopes and Radiation Technology*.[5] This structure consists of two cylindrical chambers, the upper of steel and the lower of concrete. The upper is used for such purposes as maintenance work on the lamp, while the lower houses the radioisotopic unit, which is mounted on the floor.

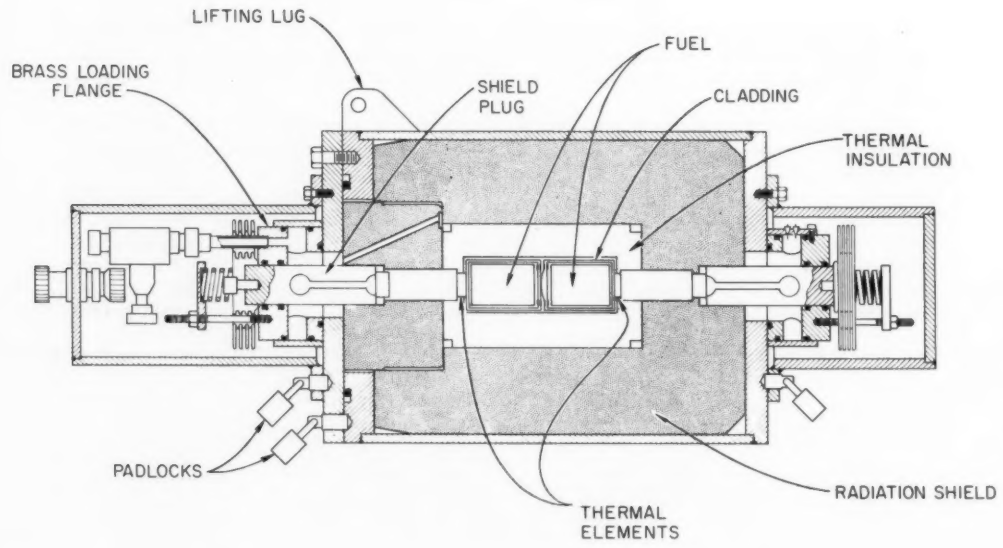

Fig. 1 RIPPLE VI radioisotopic energy source used in Sweden's radioisotope-powered lighthouse.

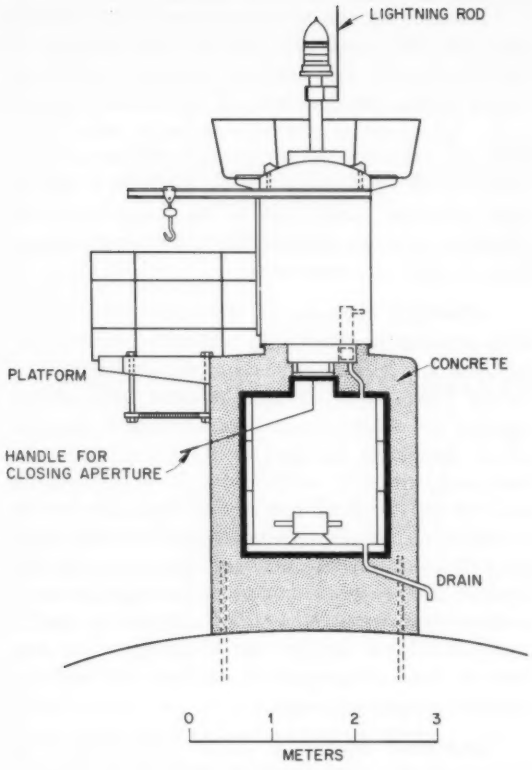

Fig. 2 The radioisotope-powered lighthouse in the Stockholm archipelago.

When the opening between the chambers is closed with its concrete cover, the radiation dose in the upper chamber is below the maximum permissible for public access.

The 10-watt lamp of the navigational unit has an operating cycle of 0.3 sec on, 1 sec off, 0.3 sec on, 4.4 sec off during the darkness hours, with an average operating time of 17 hr/day. The cycle is switched in and out photoelectrically. The generator charges two banks of Ni–Cd batteries, each of five cells, with a total capacity of 1500 watt-hr at 12 volts. A d-c to d-c unit converts the 4-volt output of the isotopic generator to 12 volts.

Only 2 days was required for installing the unit, including a 25-mile sea trip from the dock to the site.

(MG)

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Radioisotope Use in France—1966

A survey[1,2] on the use of radioisotopes in France was made in 1967 to update one made[3] in 1961–1962. The reviews[4,5] of the status of radioisotope use, made in 1966, were not based on a comprehensive survey.

For the 1967 survey, questionnaires were sent to 1351 users asking about their uses of radioisotopes in 1966. Replies were received from 967, or 72%, of those queried, of which 59% used radioisotopes and 13% did not. The results showed a 75% increase in radioisotope users between 1961 and 1966. The increase in the number of nuclear gages in use was significant, and indications were that the upward trend was continuing, as was also the use of nondestructive testing methods based on radioisotopes. Ionization, tracer, and large-scale irradiation uses appeared to be developing somewhat less rapidly, but the data were too few to be treated statistically.

The industries surveyed included rubber, plastics, textile, metal, chemical, electronic, petroleum, photographic, machine, and glass. The radiation properties on which applications were based were absorption of radiation by materials, as in thickness or level gages and radiography installations; ionizing ability, as in static eliminators, food-preservation facilities, and vacuum gages; and on the ready detectability of radiation, as in tracers. A total of 1037 uses of radioisotopes was reported, with some installations using them in more than one way:

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Type of use	No. of users
Research	118
Control	489
Fabrication	404
Other	26
	1037

Gaging. The total number of gages in use reported in the 1967 survey was about twice the number in use in 1961, viz., 2478:

Type	No. in use	Total investment, million F*	Predicted increase by 1970, %
Thickness	662	15.0	65
Level	1577	6.0	50
Density	172	4.3	75
Moisture	58	–	–
Analysis	9	–	–

*1 F = approximately $0.20.

The number was rather high even in 1961, and the market does not seem to be saturated yet. Most of the gages, with the exception of those used for analysis, are used in continuous process control. The average cost per gage, including auxiliary equipment, was 35 thousand F ($7 thousand) for thickness measurements; 7.3 thousand F ($1.4 thousand) for level; and 33.5 thousand F ($6.7 thousand) for density. Annual maintenance costs are about 10% of the gage cost in each case. Data on other types of gages were too few for meaningful cost figures to be calculated.

Radiography. A total of 461 radiography installations, for nondestructive testing, was reported—mostly gamma but a few neutron—in 171 plants. The total in use cannot be compared with the total given by the earlier survey because in 1961 the number applied to applications, while the 1967 number applied to installations.

The average cost per installation was 170 thousand F ($34 thousand), with annual maintenance calculated to be less than 2% of this value. The total investment is put at 5.7 million F ($1.14 million), and the number of installations is expected to increase 70% by 1970.

Tracers. The use of tracers showed a 41% decrease from the 1961 survey data. This was partly because the method of collecting data was different—unlike the earlier survey, the later one did not include personal visits. Also, however, the CEA no longer makes tracer tests for industry. Tracer uses reported were 43% in research, 35% in process control, and 22% in fabrication techniques. Only 38% of the tracer users took advantage of rental services. The others were equipped to make their own tracer tests.

Activation Analysis. In the 1961–1962 survey, only 16 users reported using activation analysis, with 15 using nuclear reactors and one using an accelerator. In the 1967 survey, 65 users reported using nuclear reactors and 5 using accelerators. The largest usage was in the petroleum and gas, chemical, and electronics industries, with 23% of the work for process control and the rest for laboratory-scale studies. The increase in industrial use of activation analysis is attributed to the efforts of L'Atom Industriel, who have expended considerable effort in promoting this application of radioisotopes. X-ray-fluorescence methods of analysis were introduced in the period between the two surveys, but charged-particle analysis has not yet become important in industry.

Large-Scale Irradiation. Some 56 establishments use large-scale irradiation, but 90% of these do so through outside facilities. The number of applications seems to have doubled since 1961.

Miscellaneous. Luminescence applications increased from 6 to 26, mostly for watch dials.

(Martha Gerrard)

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Determination of Wear with Radioisotopes
(Determinarea uzurii cu radioizotopi)
By Al. Chişu and A. Szabo, Editura tehnica, Bucharest, 1967

This 318-page book (105 references) in Romanian is a basic treatise on determination of wear in various machine parts with radioisotopes, used either as tracers or in thickness-type gages. It is well illustrated and contains much basic theory. (MG)

Australian Radioisotope Production and Services—FY 1968*

In its *Sixteenth Annual Report 1967–68,* the Australian Atomic Energy Commission (AAEC) indicated a 57% increase in demand for radioisotopes during the year ending June 30, 1968, as compared to FY 1967 (Table 1). The rise in industrial use was general, with a particular increase in the demand for ^{192}Ir gamma radiography sources. A demand for special ^{60}Co radiography sources also appeared during the year, after a long period of no interest in this item. A 117% increase in medical radioisotopes was due to expansion in the use of radioisotope diagnostic techniques in Australia, combined with the growth of radiopharmaceutical production in the recently established AAEC radiochemical production laboratory.

Imported radioisotope values were

1966–1967	1338 items	A$54,713
1967–1968	1058	56,578

These figures show a decrease in the number of items imported from 27% of the total in FY 1967 to 17% in FY 1968, although there was a slight increase in the dollar value.

A slight rise in export items, excluding ^{60}Co, was due to an increasing interest by New Zealand in AAEC medical radioisotopes. Four teletherapy sources were supplied by the Commission during the year—one to Japan, one to New Zealand, and two for Australian use.

The total radioisotope sales since the AAEC began production in 1960–1961 has now exceeded A$500,000, with almost half the income being derived from ^{60}Co sales.

During FY 1968 the production of ^{99}Mo/^{99m}Tc generators was established on a limited scale, and, by the end of the year, production had risen to about 60% of the present Australian demand. Other radiopharmaceuticals added to the production list include ^{197}Hg-labeled chlormerodrin and ^{18}F. The following processed radiopharmaceuticals of short and medium half-life are now available from the AAEC on a routine basis:

Radioisotope	Form	Half-Life
^{24}Na	Isotonic solution	12.5 hr
^{42}K	Isotonic solution	15 hr
^{82}Br	Isotonic solution	36 hr
^{32}P	Carrier-free solution Isotonic solution (buffered)	14 days
^{64}Cu	Isotonic acetate solution Isotonic EDTA complex	12.8 hr
^{18}F	Isotonic solution (buffered)	1.7 hr
^{197}Hg-labeled-chlormerodrin	Isotonic solution	65 hr
^{99}Mo/^{99m}Tc generators	Alumina column	^{99}Mo, 67 hr ^{99m}Tc, 6 hr

Australian short-lived-isotope production is based on nuclear reactor work, it having been shown in preliminary tests that the University of Melbourne cyclotron cannot produce radioisotopes in sufficient quantity for industrial use.

The AAEC, with advice from medical specialists, has completed a preliminary design and feasibility

*Australian use and production of radioisotopes in earlier years was reviewed in *Isotopes and Radiation Technology.*[1,2]

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Table 1 Australian AEC Production of Radioisotopes

Use	Product	1966–1967 No. of items	1966–1967 A$	1967–1968 No. of items	1967–1968 A$
Industrial	^{60}Co and ^{192}Ir radiography sources	104	16,433	156	24,816
	Misc. encapsulated sources (^{60}Co, ^{192}Ir, etc.)	95	3,070	356	1,760
	Tracers and misc. products	159	1,652	1,167	6,479
		358	21,155	1,679	33,055
Nonmedical research	^{60}Co sources	7	21,743	25	6,267
	Miscellaneous	385	7,159	782	8,438
	Service neutron irradiations	289	4,879	68	1,434
		681	33,781	875	16,139
Medical	Radioisotopes for implantation (^{182}Ta, ^{198}Au, ^{90}Y)	948	2,075	1,078	2,580
	^{99}Mo/^{99m}Tc generators and solutions of ^{24}Na, ^{42}K, ^{18}F, ^{32}P, etc.	219	3,674	402	10,424
	Teletherapy sources	2	30	2	20,706
		1,169	5,779	1,482	33,710
Other	Educational teaching sources	1,045	5,686	684	5,677
	Misc., including stock radioisotopes	2	15	4	16
		1,047	5,701	688	5,693
Export	^{192}Ir radiography sources	4	640	3	587
	^{60}Co high-specific-activity sources	34	40,050	2	13,854
	Medical implants	263	822	450	1,084
	Medical solutions	5	54	48	363
	Nonmedical research	6	148	12	213
	Service neutron irradiations	1	10		
		313	41,724	515	16,101
		3,568	108,140	5,239	104,698

study of a beta-ray skin-treatment machine in which the less-desirable low-energy particles from a ^{90}Sr source are deflected magnetically from the treatment beam. The technical feasibility was established, and a prototype unit is expected to be constructed for clinical evaluation.

Many uses of radioisotopes were discussed at the symposium[3] on industrial uses of radioisotopes held at the University of New South Wales in October 1967, sponsored by Unisearch Limited. A partial list of Australian open literature publications[4-17] indicates the scope of Australian work in this field. (MG)

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South African Radioisotope Production and Services in 1967

In its Eleventh Annual Report (pp. 23-26), for 1967, the Atomic Energy Board of the Republic of South Africa indicated a continuing development of radioisotope applications. One hundred and twenty-nine new authorizations were granted for the use of radioactive materials—71 industries, 28 universities and research institutions, 10 government departments and local authorities, 11 mines, 6 hospitals, and 3 medical practitioners. Nearly 200 nonmedical institutions were using isotopes as of December 31. Some 13.5 curies of radioisotope activity were produced, and 11 thousand curies were imported—nearly double the amount imported in 1966. The 0.770 curie exported consisted of ^{7}Be, ^{109}Cd, ^{139}Ce, ^{22}Na, and ^{85}Sr.

Among the new radioisotope applications investigated was the determination of the residence time of cement in a rotating kiln at the Hercules factory and of the mixing of powders during emptying of a silo filled with powder. At the request of two steel producers, the feasibility of identifying different kinds of steel with radioisotope tracers was demonstrated. Measurement of the Vaal River flow during overflow of the Vaal Dam was attempted, but, despite the turbulent flow, the tracers did not mix thoroughly with the water. Projects were started on accurate measurement of waterflow in cooling systems and the undesirable transfer of silicon compounds to the steam phase in boilers.

At the request of a local firm, a method for weighing ore on a moving conveyor belt using gamma-ray absorption was devised.

In the field of biology, iron- and vitamin B_{12}-absorption studies were made.

A one-day national symposium on neutron activation analysis and production of isotopes in the research reactor Safari I was attended by 130 delegates. Safari I has been commissioned and is available to outside organizations for service irradiations. A start has been made in South Africa on identification of human hair by activation analysis. (MG)

XUM

The ^{137}Cs Gamma-Irradiation Greenhouse at Kasetsart University

By A. Nakornthap*

The botanical gamma-irradiation facility at Kasetsart University is a modified greenhouse (Fig. 1) in which a 100-curie ^{137}Cs source is located. This source was installed in 1965, replacing a 16-curie ^{60}Co source used earlier. The irradiation chamber (Fig. 2) is an 8- by 10-m room, shielded on all four sides with concrete whose thickness is such that gamma dose rates outside the walls do not exceed 2 mr/hr.

The walls are high enough to prevent direct radiation from reaching an adjacent building containing offices and lecture rooms, and any gamma radiation emitted through the roof will be scattered by the air, the roof, and supporting structures above the cell. This scattered radiation, or "sky shine," is negligible in effect. The chamber is connected to an 8- by 13-m nursery, and the whole building has a glass roof.

The source, counterweighted by a 6- by 18-cm lead plug, is contained in a hollow tube of about 7 cm diameter. The tube is sunk 2.5 m in the earth, the aboveground part being of aluminum and the belowground part of stainless steel and surrounded by a flint-coated iron pipe. When the source is 2.5 m below the ground level, no radiation is detectable above the concrete floor. When in use, the source is raised in the tube to about 52 cm above the floor, and plants to be irradiated are placed around the source, at distances determined by the dose rate desired. The tube is located in the cell equidistant from the side walls, 3.35 m from the back wall, and 6.65 m from the front.

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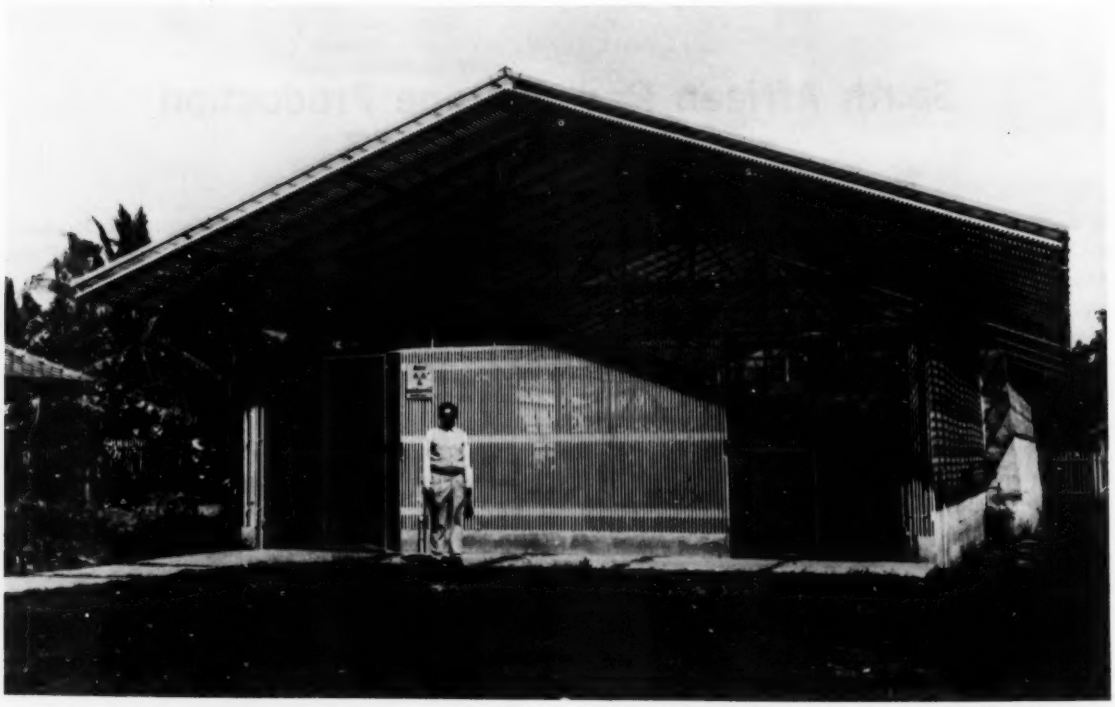

Fig. 1 Rukka Rangsee (*rukka* means "plants," *rangsee* means "radiation") greenhouse irradiation facility of Kasetsart University.

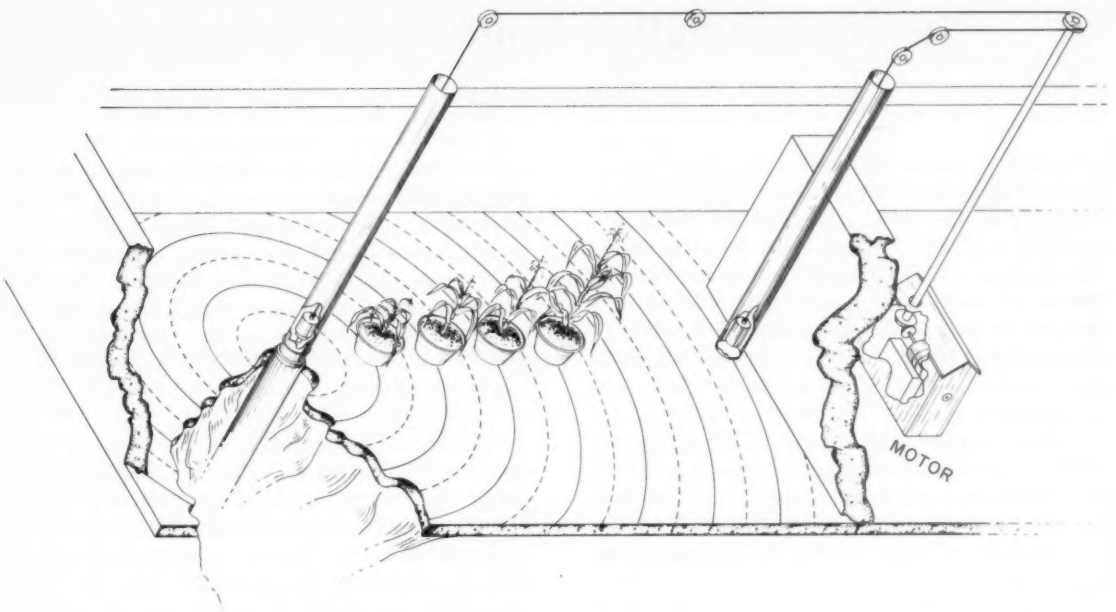

Fig. 2 Gamma-irradiation room at Kasetsart University facility, showing ^{137}Cs source in operating position.

The source is raised and lowered electrically by a motor located outside. Manual operation is possible in case of electrical failure. The source is in use 17 hr out of 24, being kept underground during most of the time that people might be around.

Safety mechanisms include a red warning light above the gamma room which goes on when the source is up. The door is always locked when the source is up, but if the door should be opened when the red light is on, a siren would be set off and the ^{137}Cs source would be automatically lowered.

The total cost of the gamma-irradiation building was about 300,000 baht (U. S. $15,000), including the 100-curie ^{137}Cs source, shipping cost, operating mechanisms, alarm systems, irradiation room, connecting nursery, and a 6- by 6-m laboratory and tool shed.

The Kasetsart facility has been used for mutation work. For example, permanent color changes have been induced in strains of canna, a popular ornamental plant in Thailand, by irradiating the rhizomes and young plants.[1] It has also been used for irradiating rice, cotton, tobacco, bean, rubber, and other seeds; bulbous and wood plant parts; fish fingerlings; insects; and other materials as well as for training purposes. (MG)

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XUM

Gamma-Irradiation Synthesis in Canada

By Sharda DasGupta*

Utilization of gamma radiation for creating new materials with desirable combinations of properties and for modifying existing industrially important products by imparting new characteristics is the basic aim of the New Materials Program at the Commercial Products Division of Atomic Energy of Canada Limited. The objective of the Canadian work is to demonstrate to industry the immense potential of ionizing radiations. Radiation-induced synthesis is still in its infancy, but the promise is immense. It is foreseen that in the not too distant future much of the synthetic work in polymers will involve radiation in some stage of the process. Research has been diversified, and processes that may be of interest to many types of industries are being worked on. Briefly, some of the more interesting projects are:

Textile Grafting. As a result of investigations of reaction mechanisms and study of changes in the molecular structure induced by gamma radiation, optimum conditions have been developed for grafting vinyl and vinylidene monomers to cotton and nylon. Grafted cotton[1-3] has remarkable fiber durability and shows high resistance to abrasion, chemicals, heat, weathering, and biological attack. While each of these properties will greatly enhance the value of cotton, the resistance to biological attack is perhaps the most beneficial characteristic because it makes cotton competitive with synthetic fibers.

Grafting on nylon[4] imparts certain specific attributes which can render it useful in new areas. For example, nylon can be made permanently waterproof and thus suitable for tenting. Other properties of interest are the increase in soil resistance and tensile strength and the marked decrease in the capacity to accumulate static charges. These properties could be of great interest in the carpet industry.

Simple procedures have been developed for the radiation grafting of hydrophilic groups such as amides and acids onto terylene[5,6] and polypropylene.[6] This affects the antistatic properties, moisture regain, and dyeing characteristics. The latter is of great importance to the textile and carpet industry because at present very complex procedures—e.g., dyeing by melting, complexing with metal, and blending with other readily dyeable fabrics such as cotton—are being used to overcome the shortcomings of these "miracle" fibers. A considerable improvement in dyeability is attained as a result of grafting. Plans are under way to prepare grafted samples of textiles and fibers large enough for industrial evaluation.

Wood–Polymer Composites. Research has been carried out to determine properties of various wood–plastic combinations produced by radiation polymerization[7] for use in improving surface and mechanical properties of wood–polymers made from Canadian soft and hardwoods. Polymerization rates have been investigated for various monomer systems together with the characteristics of the polymers formed, and data have been obtained for use in controlling both the quantity of monomer absorbed by various wood species and the temperature–dose rate effects.[8] Plans are being made for producing large sheets with improved surface properties.

Grafted Nylon Molding Powders. There is a great demand in the consumer market for materials that have good molding characteristics, combined with inertness and ability to perform at high temperatures. Considerable progress has been made in our laboratories in the synthesis of such materials. Grafting of the ladder polymer polynaphthylidine-acrylonitrile onto commercial nylon[9] molding powder yields a product with high resistance to heat and enhanced inertness to radiation, chemicals, and common solvents. The grafted copolymer when heated to 450°C loses only 5 to 8 wt.%, while the corresponding value for untreated nylon is 90%. Styrene-grafted nylon[10] powders have also been synthesized. These show high resistance to moisture, chemicals, and radiation and have excellent molding characteristics.

The cost for carrying out these important modifications is likely to be very small. We estimate that grafted nylon molding powder prepared on a large scale would cost no more than the ungrafted powder since the increase in weight achieved by grafting would compensate for the cost of the procedure, styrene being relatively cheap.

*Atomic Energy of Canada Limited, Commercial Products, P. O. Box 93, Ottawa. This paper was presented at the first USAEC–AECL information-exchange meeting, Brookhaven National Laboratory, December 1968, under the title "New Materials Program."

Grafting on Paper. Methods have been found for grafting vinyl monomers onto newsprints, kraft, and sulfite paper.[11] Several variables have been studied and a variety of grafted samples obtained. The products show 200 to 300% increase in wet mechanical strength, and they resist moisture, rot, grease, and oil. This expands the scope of use for paper in the packaging industry. Styrene- and acrylonitrile-grafted papers have been shown to render surfaces more suitable for printing—a factor of considerable interest for the production of newsprint.

Radiation-Resistant Lubricants.[1] Lubricating agents that have hydrocarbon molecules, such as greases and oils, undergo changes when irradiated[12] to doses of 10 to 15 Mrads. These doses not only destroy the lubricating properties to a very great extent but cause formation of acids that may be corrosive. In any location where intense radiation fields are encountered, there is great need for radiation-resistant greases. Many forms of solid lubricating films, though relatively immune to the action of radiation, suffer from drawbacks such as flaking due to vibration and other limitations in their lubricating properties.

We have conducted work with a variety of well-known high-grade commercial greases with the aim of inducing radiation resistance. Styrene monomer has been grafted to hydrocarbon-base lubricants through the use of gamma radiation. The grease copolymers were formed without destruction or change in any of the physical characteristics of the starting material. It must be stressed that this would not have been possible with conventional chemical procedures. The new products showed many performance characteristics that were improved significantly over those of the corresponding starting materials. However, the most important feature was their radiation resistance. When irradiated to high dose levels—up to 10^9 rads—samples of the grafted lubricants showed no deterioration as compared to unirradiated commercial greases. It has been established that the new product can absorb 250 to 1000 times as much radiation as the starting material before the same degree of deterioration occurs.

Modification of Lignin. Lignin graft copolymers with aldehydes and vinyl monomers have been synthesized using radiation catalysis. The optimum conditions for preparing such copolymers have been established and the various reaction mechanisms investigated. Lignin may be considered a colloidal thermoplastic polymer made up of cross-linked polyaromatic chains with propyl side branches. Since the chains have high radiation resistance, proper conditions had to be found for grafting to occur on the propyl branches with low radiation doses. Such copolymers have been found to be remarkably useful as dye dispersants, ore flocculants, etc.

Other Projects. We are also currently carrying out exploratory research on a variety of other possible applications, e.g., the radiation-induced synthesis of polyorganosiloxanes. Inorganic polymers are complex structures that do not contain carbon in the primary molecular chain, e.g., polyorganic siloxanes and polymetalorganic siloxanes, in which silicon replaces carbon, and various atoms—Al, B, Ti, Sr, Co, Ni, etc.—form part of the complex. Such polymers show remarkable stability even at high temperatures. For example, polytitanomethyl siloxane does not flow or become oxidized even at temperatures up to 700°C.

The characteristics of high resistance to heat and relative inertness obviously limit the application of conventional chemical procedures for the synthesis or modification of these materials. The use of radiation-induced reactions is therefore being looked into for obtaining a larger variety of such unique molecules. Initial results are most encouraging. (MG)

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10. S. DasGupta, Creation of Polymeric Structures for Space Material Development, *Can. Aeronaut. Space J.*, **11**(5): 133-141 (1965) (AECL-2172); *Gamma Irradiat. Can.*, **4**: 103-120 (1965) (AECL-2298). (Presented at the Canadian Aeronautics & Space Symposium, Toronto, February 1965.)
11. S. DasGupta, Use of Co-60 for the Production of Industrially Important Materials (AECL-1789). (Presented at the International Canadian Nuclear Association Conference, Montreal, May 1963.)
12. S. DasGupta and J. T. Slobodian, New Greases Withstand High Radiation Doses, *Can. Nucl. Tech.* (Fall 1963) (AECL-1832). (Presented at the 142nd American Chemical Society Meeting, Atlantic City, September 1962.)

Isotopes in Chemistry
By J. F. Duncan and G. B. Cook
Clarendon Press, Oxford, 1968 (258 + xv pp.; $6.90, paperback)

The authors of *Isotopes in Chemistry* feel that emphasis on the convenience and uniqueness of isotopic methods sometimes obscures the broader possibilities of application of such methods. They therefore have attempted to show how isotopic methods of investigation can be used to study fundamentals of chemical systems. In support of this effort they have included chapters on isotope effects and on chemistry of nuclear processes, with sections on, e.g., radioactive recoil, the Mössbauer effect, and nuclear magnetic resonance. Tracer methods are not neglected; for example, radioactive reagents, isotope dilution, and radiometric titration are covered. Nontracer isotopic methods described are activation analysis, absorptiometry, and X-ray fluorescence. The final chapter is on the chemistry of the radioactive elements.

The authors have made no attempt to write an exhaustive treatise. Rather, they have presented typical situations in a textbook designed for advanced undergraduate students. The book is well documented, but, as in most textbooks, the dates of the references lag about 2 years behind the publication date. (MG)

Kyoto University Gamma-Irradiation Facility for Biochemical Research

By Sakae Shimizu and Hiroyuki Hatano*

Three interesting features of the ^{60}Co gamma-irradiation facility (Fig. 1) used in radiation-biochemistry work at the Kyoto University Chemistry Department are a system for controlling the temperature anywhere between −25 and +95°C, a sample holder through which liquid samples can be circulated for continuous irradiation, and a system for automatic chemical measurements during an irradiation without interruption of the irradiation. This last system is particularly useful when unstable intermediate products must be isolated and determined immediately.

The 100-curie ^{60}Co source can deliver doses at rates varying from 6×10^3 to 5×10^4 r/hr. It is made up of 10 pellets of 10-curie/g cobalt, each pellet being 5.2 mm in diameter by 7.5 mm thick. The 10-pellet unit is doubly encapsulated in stainless steel, an inner cylinder of 5.5 mm diameter by 75 mm high and an outer of 8 mm diameter by 90 mm high. The source is housed in a cylindrical shield equipped with a shutter. When the shutter is closed, radiation at the outer surface of the housing is less than 2 mr/hr. The source and housing are mounted on the end of a rod that moves up and down in a vertical shaft that opens into

*Radioisotope Research Laboratory, Institute for Chemical Research, and Chemistry Department of Kyoto University, Kyoto, Japan.

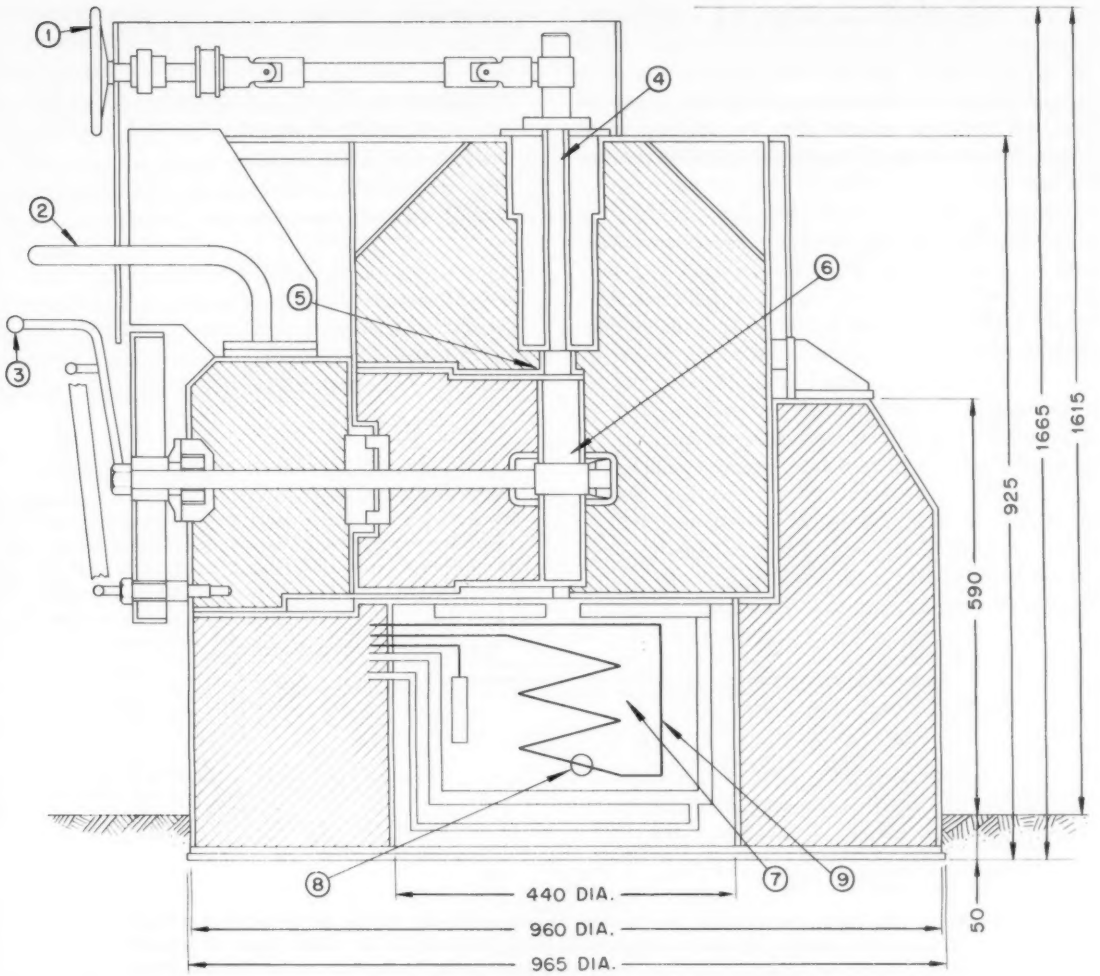

Fig. 1 Kyoto University gamma-irradiation facility for biochemical research: (1), (2), and (3), handles for up–down movement of radioactive source, container movement, and shutter manipulation, respectively; (4) rod for moving source capsule; (5) source capsule; (6) shutter; (7) irradiation chamber; (8) source in irradiation position; (9) circulating-sample pipe. Shading on diagram represents lead shielding. Dimensions are in centimeters.

the top of the irradiation chamber, and the source is kept in the shaft when not in use.

The irradiation chamber is a stainless-steel cylinder 30 cm in diameter by 20 cm high. Around it is a spiral copper tube—32 cm in diameter by 22 cm high—through which liquids can be circulated to regulate the chamber temperature. The whole arrangement is insulated with glass wool. The shaft and chamber are shielded by lead. When an irradiation is to be carried out, the sample is placed in the irradiation cell, and the source is lowered into the chamber and manipulated until it is located at the position where it gives the desired dose rate. The door to the chamber is closed, and the shutter on the source housing is opened. A mechanical interlock system prevents opening of the shutter until the irradiation-chamber door has been closed. When the irradiation is finished, the reverse operations are carried out. The source, sample, and shutter are manually moved about inside the cell by means of handles outside the cell.

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The circulating-sample holder is a stainless-steel spiral pipe—25 cm in diameter by 18 cm high—made of 4-mm-diameter tubing. The system (Fig. 2) is equipped with a 15-watt pump, which can deliver 3.5 liters of liquid per minute. The combination of an autoanalyzer with this system makes possible continuous study of chemical reactions and determination of enzyme activities under the influence of gamma radiation. The stationary sample holder is a stainless-steel test-tube stand 29.7 cm in diameter by 14.5 cm high. It can accommodate 24 sample tubes of 18 mm diameter or 33 of 12 or 5 mm diameter. (MG)

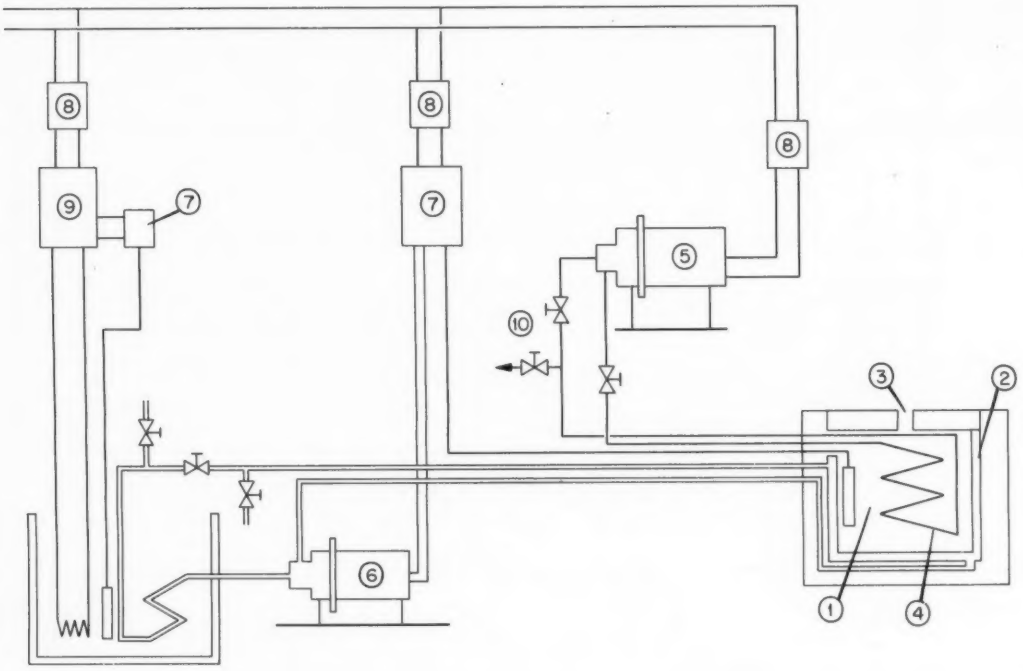

Fig. 2 Circulation systems of Kyoto University gamma-irradiation facility for biochemical research: (1) irradiation chamber; (2) constant-temperature bath; (3) opening for in–out movement of radiation source; (4) circulating-sample pipe; (5) circulating-sample pump; (6) constant-temperature bath pump; (7) thermostat; (8) switch; (9) magnetic switch; (10) outlet to autoanalyzer.

Korean ^{192}Ir Radiography Handbook

A 170-page handbook* for guidance of manufacturers who use ^{192}Ir gamma radiography in nondestructive testing has been issued by the Korean Atomic Energy Research Institute. A few Korean manufacturers have started to use gamma radiation in nondestructive testing, but no codes and standards for radiography had been established in Korea and no guidebooks were available in Korean prior to issuance of this report. The report therefore gives general concepts and procedures required for nondestructive testing by radiography and information required for evaluation of radiographs. Particular emphasis is placed on the use of ^{192}Ir, which is available from the Korean TRIGA reactor. (MG)

*Kwan Lee and Won Guk Hwang, *Gamma Ray Radiography by Ir-192* (in Korean), Atomic Energy Research Institute, Seoul, Korea, Report AERI-21, 1968.

Statistics of Radiation Use in Japan—1968*

The Japan Radioisotope Association has published a 31-page brochure, in Japanese and English, giving statistics on the use of radioisotopes in Japan for the period March 1959 through March 1968. Information on the amounts of radioactive wastes disposed of is included.

The total number of establishments licensed to use radiation increased from 50 in March 1959 to 1540 in March 1968 (the Japanese fiscal year ends in March), with the greatest percentage increase by hospitals and industry (Fig. 1). These numbers include both radioisotope sources and cyclotrons or accelerators, but not X-ray machines. A categorization of the 1968 total shows 1349 establishments licensed to use sealed sources, 408 to use nonsealed sources (tracers), and 127 to use radiation generators. The discrepancy between the total of these values and 1540 is due, of course, to some establishments' being licensed for more than one kind of source. Breakdowns of the amounts of the individual nuclides distributed for tracer use and for sealed-source use, both large sources and in gages, etc., are given in Tables 1 and 2.

The number of organizations in the business of making and handling radioisotopes and related equipment has increased steadily but may be leveling off (Table 3).

The number of persons trained in the use of radioisotopes increased from 64 in 1957 to 323 in 1967. Certificates were issued in 1967 to persons trained to handle sealed sources; to perform autoradi-

*Published by the Japan Radioisotope Association, 28-45 Honkomagome 2-chome, Bunkyo-ku, Tokyo 113. Information from this brochure and from other sources was also presented by I. Suetsuna in "Present State of Development and Utilization of Radioisotopes and Radiation in Japan—with Emphasis on Their Industrial Utilization," *Nucl. Eng. (London)*, **14**: (1969).

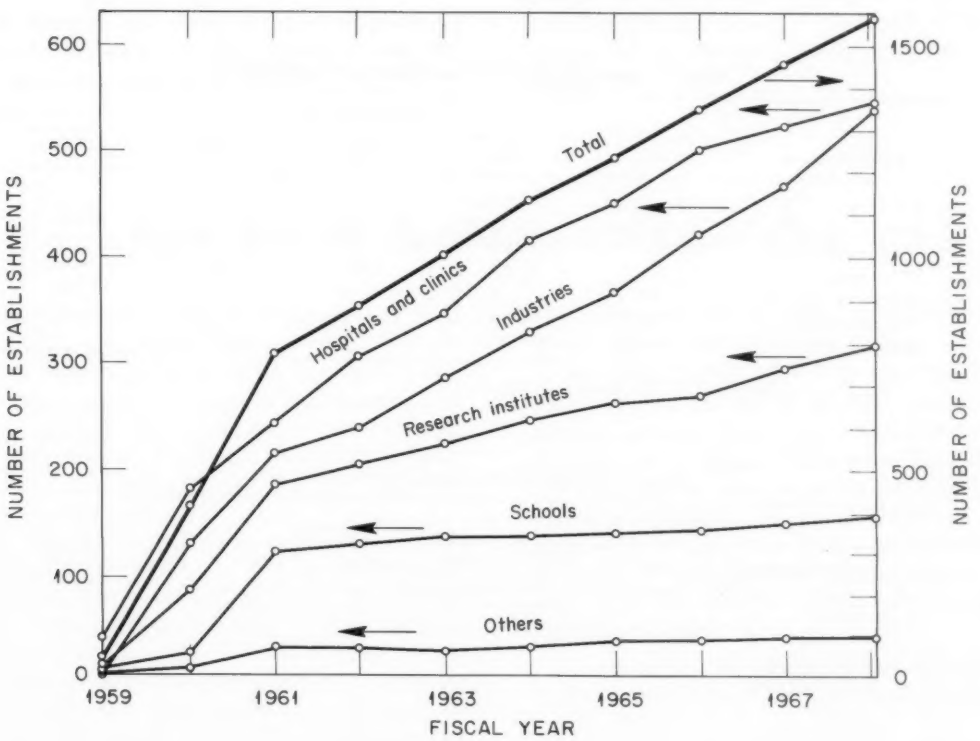

Fig. 1 Increase in number of establishments using radiation from 1959 to 1967.

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Table 1 Amounts of Nonsealed Radioisotopes Distributed

Nuclide	Amount, mc				
	1963	1964	1965	1966	1967
^{3}H	2,450,000	1,060,000	4,740,000	6,800,000	4,040,000
^{14}C	580	1,030	760	1,090	1,850
^{24}Na	230	680	510	400	240
^{32}P	32,300	26,900	30,000	27,700	25,100
^{35}S	3,100	2,470	3,400	3,130	2,300
^{45}Ca	420	320	900	350	490
^{51}Cr	1,240	980	1,040	950	2,900
^{59}Fe	240	310	270	240	340
^{57}Co	70	50	40	50	50
^{75}Se	20	30	80	240	370
^{85}Kr	700,000	350,000	550,000	360,000	770,000
^{85}Sr	50	50	30	80	160
^{99m}Tc	–	–	2,150	8,300	9,700
^{125}I	80	260	590	850	580
^{131}I	61,000	63,100	70,700	72,500	69,000
^{133}Xe	100	1,600	2,500	2,700	4,220
^{137}Cs	190	140	150	1,760	2,630
^{147}Pm	1,370,000	1,430,000	1,600,000	4,510,000	7,300,000
^{198}Au	38,700	35,900	34,400	47,800	50,200
^{203}Hg	380	430	790	1,710	1,450
Others	–	–	–	–	3,170

Table 2 Amounts of Sealed Radioisotopes Distributed

Kind of source	Amount, curies				
	1963	1964	1965	1966	1967
α sources	0.02	0.68	0.62	0.67	0.49
β sources	7.1	11.7	63.0	58.7	10,098*
^{60}Co large sources	173,000	219,000	234,000	89,700	102,800
^{137}Cs large sources	1,200	8,100	4,000	5,200	670
Other large sources	60	38	44	177	1,500
Neutron sources	20.5	6.1	27.6	30.0	34.6
Other sources	–	23.4	1.8	450.4	6.4

*Includes 10,000 curies of ^{90}Sr for heat sources.

Table 3 Organizations Handling Radioisotopes

Dealer category	Number of dealers						
	1962	1963	1964	1965	1966	1967	1968
Producers	–	1	1	3	3	3	3
Source processors	1	1	6	6	8	9	10
Instrument and equipment manufacturers	–	–	5	10	15	19	19
Traders	–	–	–	–	2	12	13
Total	1	2	12	19	28	43	45

ography; to carry out activation analyses and radiation chemistry; and to apply radioisotopes in biology and in industry.

The brochure contains a total of 42 tables and 17 figures, which give details of various kinds, e.g., radioisotope utilization by type of industry; radioisotope distribution by local area in Japan; waste collection by district; change in the number of X-ray machines used; number of trainees by type.

(MG)

New French Isotope Catalog

A 1968 catalog, *Radioéléments et Molecules Marquées* (Radioactive Elements and Labeled Molecules), issued by the French Commissariat à l'Énergie Atomique—Centre d'Études Nucléaires de Saclay (Mol–SORIN) replaces the 1966 edition.

Besides listing sales items, the catalog includes many technical details. For 104 radioisotopes (63 elements) are cited the half-lives; reaction cross sections and target compositions used in their preparation; and activities of the products obtained after 7 days, 30 days, and saturation irradiation with a thermal-neutron flux of 10^{13} n/(cm^2)(sec). The common radioactive elements are tabulated by half-life and radiation energy.

One hundred and thirty-six radioisotope-labeled compounds (88 elements) for nonmedical usage are listed with the radioisotope half-lives and specific activities. A radiopharmaceutical list also includes product sterility data. More than 450 materials labeled with ^{14}C, ^{32}P, ^{35}S, and ^{3}H supplement the list in the old catalog, and customers may send their own compounds to be tritiated by the Wilzbach or exchange process or by addition to unsaturated precursors.

New sealed sources available are ^{241}Am for alpha; ^{152,154}Eu, ^{109}Cd, ^{125}I, and ^{169}Yb for gamma; and ^{241}Am–Be for neutrons. Nonsealed solid sources are ^{210}Po and ^{241}Am for alpha; ^{85}Kr or ^{3}H adsorbed on a metal for beta; and ^{3}H bremsstrahlung sources. Solid implantation sources for therapy include, for the first time, ^{32}P. Yttrium-90 granules are offered in five sizes instead of just three as formerly, and ^{192}Ir is offered as wire and pins as well as spheres. Generators for short-lived radioisotopes—^{99m}Tc and ^{113m}In—are included for the first time, and industrial ^{60}Co sources are described.

(MG)

New British Radioisotopes Catalog

More than 2000 items are listed in the new edition of the Radiochemical Centre's catalog, *Radioactive Products.* The 140 new products and services, together with many improvements in product specifications and delivery, make this the most comprehensive range of radioactive materials available from a single supplier. The catalog itself was produced in a new format for easier reference by the user.

The 120 new labeled compounds include amino acids and carbohydrates as well as more-complex biochemicals. Improvements have been made in the specific activity of tritium- and ^{14}C-labeled compounds, and many of the latter are now available at levels approaching the theoretical maximum. Standardized pack sizes with fixed activities on specific dates have been introduced to facilitate predispensing and stockholding and permit higher standards of quality control, quicker delivery, and larger decay allowances.

The newly introduced ^{242}Cm alpha and neutron sources are on the list of industrial and laboratory radiation sources, which also features considerable extensions to the ranges of Mössbauer, beta, positron, and low-energy gamma sources and targets for neutron generators. Additions to clinical radiation sources include sphere-type ^{60}Co ophthalmic applicators for treatment of retinoblastoma and extensions to the range of ^{137}Cs tubes for intracavitary radiotherapy and for afterloading.

Copies of the catalog are available free from The Radiochemical Centre, Amersham, Bucks, England.

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20 Years of Production and Application of Isotopes and Nuclear Radiation Sources in the National Economy of the USSR

The USSR National Committees for the Use of Atomic Energy and of Soviet Ministers for Science and Technology together with the USSR Academy of Sciences held a conference in Minsk on Oct. 7–12, 1968. The conference featured papers on production and uses of isotopes and nuclear sources of radiation in the USSR and marked the 20th anniversary of the first production of radioisotopes in that country. The Russian atomic energy journal for February 1969 (*Atomnaya Energiya*, Vol. 26, No. 2) is devoted entirely to a presentation of the plenary papers and reviews of sectional sessions of the conference. The table of contents of this issue is:

Introduction

Isotopes in the National Economy of the USSR, A. M. Petrosyants 100

Plenary Papers

Progress in Production of Isotopes, V. V. Bochkarev, N. E. Brezhneva, and E. E. Kulish 106
Isotopes in Scientific Investigation, V. I. Spitsyn 114
Isotopes in Industry, E. E. Kulish and A. S. Shtan' 122
Nuclear Methods of Analysis, I. P. Alimarin and Yu. V. Yakovlev 127
Radioactive Isotopes for Diagnosis and Treatment of Diseases of Different Human Organs and Systems, G. A. Zedgenidze 133
Synthesis and Investigation of New Isotopes and Elements, G. N. Flerov 138
Mössbauer Effect Applications to Science and Technique, V. I. Gol'danskii 142
Radiochemical Processes in Industry, V. L. Karpov 150
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Results of Using Isotopes and Radiation in Research on Agriculture, S. V. Nerpin, V. M. Prohorov, and V. N. Savin 161
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Radioisotopic Thermoelectric Generators, G. M. Fradkin and V. M. Kodyukov 169

Reviews of Sectional Sessions

Radioisotopes in Monitoring Regulation and Automation of Technological Processes in Industry (B. P. Bulatov, V. A. Yanushkovskii) 176
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Nuclear Physical Methods for Materials Analysis (I. P. Alimarin, B. G. Egiazarov, G. I. Kir'yanov, Yu. V. Yakovlev, A. L. Yakubovich) 201
Nuclear Geophysics (M. G. Evteev, V. I. Ferronskii) 204
Production of Isotopes (V. V. Bochkarev) 207
Tracer Methods in Research and Industrial Production (P. L. Gruzin, Yu. F. Babikova) 212

(MG)

Canadian Biological Irradiator

Atomic Energy of Canada Limited, Commercial Products Division, has announced the availability of its Gammacell 20 for irradiation of small animals and biological samples. The unit, which weighs 6500 lb and has overall dimensions of 97.5 by 45 by 28 in., contains two doubly encapsulated ^{137}Cs sources, 1000 curies each. The unit is lead-shielded, with provision for housing the sources in the shield. The irradiation chamber has a 222-cu in. volume ($1\frac{7}{8}$ in. deep by 12 in. in diameter). (MG)

Wear in Floor Covering Detected with Radioisotopes*

A nondestructive method developed at the TNO Central Laboratory in Delft, Netherlands, for determining wear in a floor covering is based on the X-ray fluorescence produced by a 1.3-mc ^{241}Am source. The fluorescence is excited in silver foils, which are placed under the rubber flooring tiles. When the ^{241}Am probe is passed over the tiles, an X ray is induced in the silver, the intensity of which depends on the thickness of the tile. A single-channel pulse-height analyzer records the X radiation, and the amount of wear that has occurred in the flooring is found by comparing the recorded X-ray signal with a calibration graph already prepared. (MG)

*A. Kemper, Wear Measurements by Means of Gamma-X-Fluorescence, *Wear*, **12**: 55-68 (1968) (Report EUR-3627*e* 1967).

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Section

VII

Isotopes and Radiation Technology

Miscellaneous

Summary of Replies to Reader Interest Check Sheet

By F. E. McKinney and P. S. Baker

The Editors of *Isotopes and Radiation Technology*, in the belief that responders to the Reader Interest Check Sheet (Fig. 1) [5(2): 167 (Winter 1967–1968)] may be interested in the results, present a summary of the replies here. Readers were asked to give their opinions of the Technical Progress Review. The questionnaire was returned to the Isotopes Information Center by 57 readers from 18 states, the District of Columbia, and 4 foreign countries. One reader pointed out that the Reader Interest Check Sheet was "hidden" at the end of the issue; perhaps this summary will call it to the attention of additional readers who would like to express an opinion.

There were five "yes or no" questions (Nos. 1, 4, 5, 6, and 7). The answers to these five questions are listed in Table 1. Question No. 2 asked how the reader received his copy of *Isotopes and Radiation Technology*. Individual subscriptions (from the U. S. Government Printing Office) accounted for 27, AEC distribution to contractors accounted for 23, and company libraries accounted for 7.

Two questions of considerable interest to the Editors were "What areas would you like to have included?" and "Do you have suggestions that might improve this Review?". Additional coverage of various subjects was suggested by 12 readers. The subjects included low-energy photon detectors, practical health physics, medical applications (three requests), industrial radiography, industrial applications, radioecology, activation analysis, and economics of using radioisotopes. One reader wanted fuller coverage of the research efforts at Pacific Northwest Laboratory that are funded by AEC's Division of Isotopes Development (DID). Another reader desired a current index of who is doing what and where, complete with subject index. A collection of engineering drawings for small colleges that would enable them to construct irradiation facilities was thought to be beneficial by one reader. One suggestion was that each issue of *Isotopes and Radiation Technology* feature an article on a radioisotope of particular current interest.

Suggestions for improving this Technical Progress Review came from 22 readers. At least 3 readers commented on the coverage given to DID contracts: abstracts with current-fiscal-year authorization, more data, and an editorial on DID's plans and goals were asked for. The style and format of the magazine itself came in for some remarks: one reader wanted it provided with punch holes for a three-ring binder, another urged more consistency in units, and one suggested that authors be listed with the titles in the table of contents. Monthly publication was urged as a measure to improve the timeliness of articles. Two readers pointed out that news items in the Miscellaneous Section are sometimes dated; on the other hand, one of these readers checked the Miscellaneous Section as being one of the most interesting sections. A reader suggested that the Isotope Technology Development Section be divided into smaller sections; it is usually the largest section in each issue. Wider circulation to management was stressed by two readers, one of whom wanted the management section to be detachable. Editorial comment on significant reports was desired by one reader. The review articles themselves were commented on as follows: experts in the field should prepare them rather than the staff of the Isotopes Information Center (a proposal with which the Editors and staff are in full accord), more theory should be included in them, and original articles should be solicited. Specific subjects were listed under suggestions for improvement as well as under suggestions for additions. These included a yearly article on isotope

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READER INTEREST CHECK SHEET

Fourteen issues of *Isotopes and Radiation Technology* have been published in the last 5 years. The editors would like your help in making this a better review and would appreciate your suggestions for improvement.

1. Do you read *Isotopes and Radiation Technology* regularly? Yes_____ No_____

2. How do you receive it? GPO subscription_____ AEC distribution_____ Library_____

3. Check the sections that are most and least interesting to you:

	Most	Least
Isotope Production and Development		
Isotope Technology Development		
Food Irradiation		
Process Radiation Development		
Source Development		
Isotopic Power Development		
Applications in Life Sciences		
Safety		
Isotopes Around the World		
Miscellaneous		

4. Are we covering your field of interest? Yes_____ No_____

   a. If not, what area(s) would you like to have included?_____

Fold

5. Is the coverage of foreign developments adequate? Yes_____ No_____

6. Is our reporting sufficiently technical? Yes_____ No_____

7. Is the bibliography in the Miscellaneous Section of use to you? Yes_____ No_____

8. Do you have suggestions that might improve this Review?_____

Please fold, staple, and return to address printed on reverse side.

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Fig. 1 The questionnaire.

Place
Stamp
Here

Isotopes Information Center
Bldg. 3047
P. O. Box X
Oak Ridge, Tennessee 37830

Staple here

Table 1 Answers to "Yes or No" Questions of the Questionnaire

Question	Yes	No	Not checked
1. Do you read *Isotopes and Radiation Technology* regularly?	56	1	0
4. Are we covering your field of interest?	50	5	2
5. Is the coverage of foreign developments adequate?	46	5	6
6. Is our reporting sufficiently technical?	53	3	1
7. Is the bibliography in the Miscellaneous Section of use to you?	47	6	4

availability and vendors, the use of isotopes in oceans, effects of radiation on dormancy of seeds, a comprehensive report on information centers, cyclotron production information, technical information on costs, and a schedule for Food and Drug Administration approval of irradiated foods.

The third question on the Reader Interest Check Sheet requested a ranking of the sections in the publication by the interest of the reader. The results are given in Figs. 2 and 3. The most interesting section by far is evidently Isotope Technology Development; the least interesting section is Isotopic Power Development. Reader preferences were consistent, i.e., the section receiving the most rankings as least interesting also received the least rankings as most interesting.

Isotopes and Radiation Technology has a circulation of about 2200. Subscriptions to the U. S. Government Printing Office account for about 1000, and the AEC distributes about 1200 free. A return of 57 Reader Interest Check Sheets—less than 3% of the readers—is somewhat disappointing. Perhaps this summary will encourage more readers to voice to the Editors their preferences and opinions.

The Editors would like to thank all the readers who took the trouble to fill out the questionnaire. We, of course, appreciate the comments of the 16 readers who

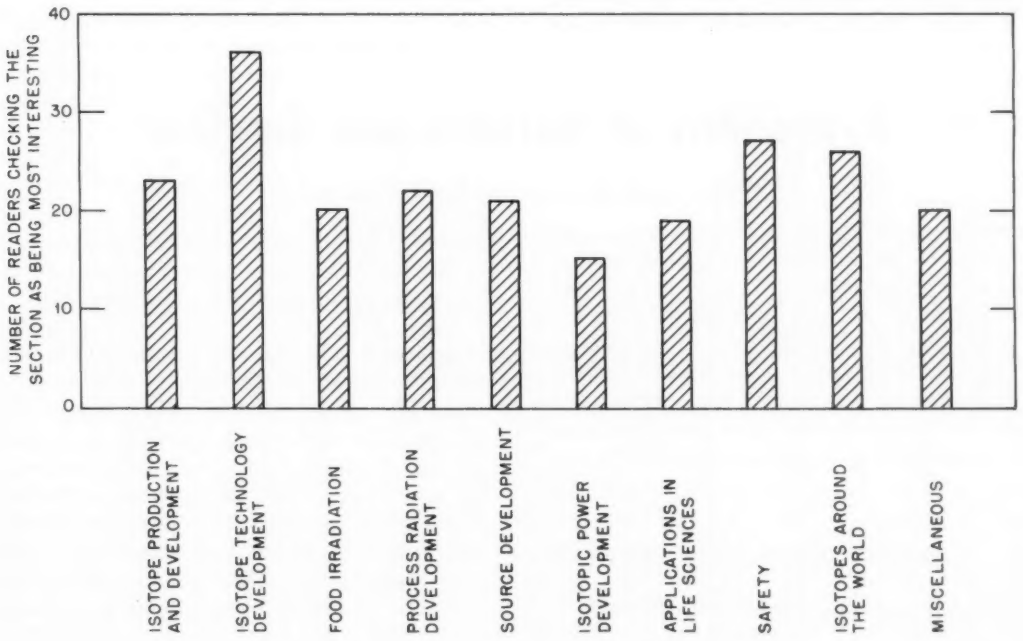

Fig. 2 Reader assessment of the MOST interesting sections in *Isotopes and Radiation Technology*.

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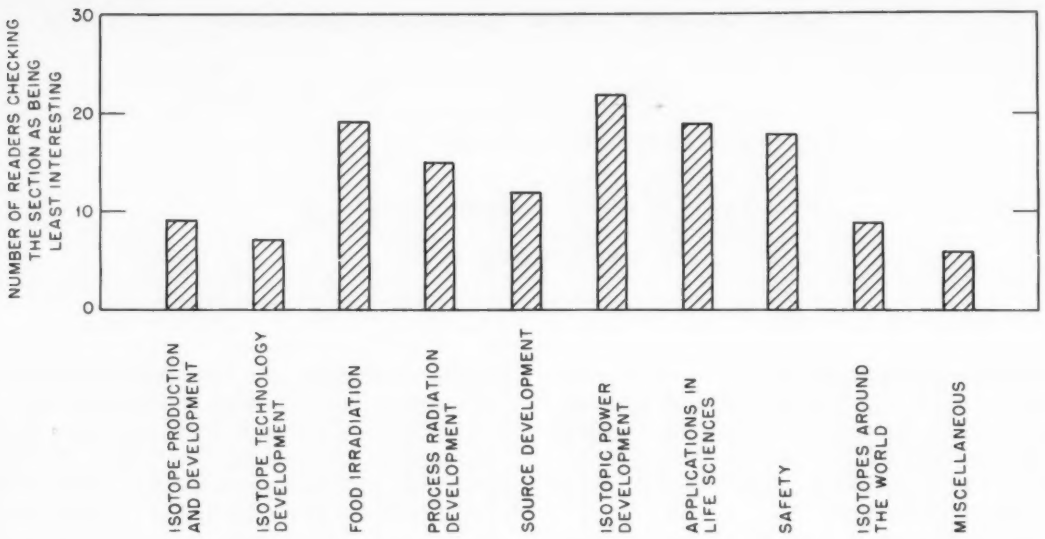

Fig. 3 Reader assessment of the LEAST interesting sections in *Isotopes and Radiation Technology*.

were complimentary about our efforts: "All is lovely," "an extremely useful review," "most interesting," "an extremely valuable service," "a good job in covering a wide range of subjects," "an excellent source for new and interesting developments," and "exceptionally good." However, we also appreciate the thoughts of those who suggested improvements and additions. In any case, the Editors will give careful consideration to the suggestions that have been made and will implement those which are feasible.

Availability of Isotopes and Services

ORNL Stable-Isotope Inventory Augmented

Recent additions to the Oak Ridge National Laboratory (ORNL) enriched-stable-isotope saleable inventory include the following:

Isotope	Purity, %	Amount available, g	Isotope	Purity, %	Amount available, g	Isotope	Purity, %	Amount available, g
^{28}Si	99.4	30.0	^{110}Pd	96.98	1.8	^{154}Gd	66.9	1.5
^{37}Cl	96.0	1.0	^{114}Sn	60.6	1.5	^{163}Dy	86.2	4.9
^{47}Ti	70.0	2.1	^{117}Sn	89.2	1.5	^{180}Hf	93.9	1.0
^{57}Fe	90.7	23.4	^{122}Te	73.3	0.4	^{196}Hg	5.1	0.3
^{64}Zn	97.8	41.3	^{122}Te	90.9	0.9	^{205}Tl	98.8	48.7
^{96}Ru	94.9	0.6	^{123}Te	77.0	0.6	^{204}Pb	70.9	25.0
^{102}Ru	96.9	3.9	^{137}Ba	81.9	6.3	^{207}Pb	84.7	311.4
^{102}Pd	75.4	0.5	^{138}La	20.0	0.06	^{207}Pb	98.2	0.9
^{104}Pd	89.7	2.2	^{146}Nd	97.4	20.0	^{207}Pb	99.8	10.3
^{105}Pd	94.5	3.8	^{148}Sm	99.9	0.015			
^{106}Pd	96.6	3.3	^{154}Sm	98.3	50.7			
^{108}Pd	98.1	2.8	^{154}Sm	98.7	48.7			

AEC Activities

^{252}Cf To Monitor Sulfur Content of Coal

The transuranium element ^{252}Cf will be used as a neutron source at the U. S. Bureau of Mines' Morgantown, W. Va., research center for activation analysis. By this technique the sulfur content of coal will be determined continuously as the coal is moving on a conveyor belt. Washing operations can then be controlled more precisely.

An on-line neutron activation instrument with a ^{124}Sb–Be source was announced earlier [*Isotopes and Radiation Technology*, **6**(4): 444 (Summer 1969)].

Large Order for ^{90}Sr Filled

The AEC's Oak Ridge National Laboratory has just filled its largest single order for ^{90}Sr—1 million curies of radioactivity. Isotopes, Nuclear Systems Division, at Middle River, Md., is to use the material for fueling radioisotope-powered electric generators.

Changes in Regulations Concerning Authority in Agreement States

The AEC has amended its regulations to help ensure uniform standards for regulating the transfer by manufacturers of products containing radioactive material for which the ultimate users are not required to be licensed. The amendment applies to products manufactured in states to which the Commission has transferred part of its licensing authority by formal agreement. Under the amendment, the Commission has authority over the transfer, by manufacturers to distributors or users, of all products that have been exempted from further regulatory control instead of only those "intended for use by the general public," as is now the case.

The use of a number of products containing small amounts of radioactive material has been exempted by the Commission from licensing control. To ensure uniform safety standards, such products must conform to conditions detailed in a specific license obtained by the manufacturer from the AEC. However, as the number of industrial proposals for exempt products increases, the Commission believes it will become more difficult to determine with certainty whether or not a product will be used by the general public. There is no technical basis for the distinction, and in some cases a device originally intended for industrial or commercial use may later be used by the public.

SNAP Generators

Two SNAP-19 generators—fueled with ^{238}Pu—are providing part of the power for the Nimbus B2 weather satellite launched April 13. They represent the first civilian use of nuclear energy in space. "Prolonged operation of this nuclear system could give satellites longer and more useful lifetimes than are possible with other systems," said AEC Chairman Glenn T. Seaborg, "and the continued success should demonstrate the value of nuclear power for missions to the moon and beyond." The Nuclear Systems Division of Isotopes, Inc., designed, developed, and fabricated the generators; Mound Laboratory, operated by Monsanto Research Corp. for the AEC, fabricated the raw fuel into the final fuel form and encapsulated it; Sandia Corp., a subsidiary of Western Electric, provided technical direction for the SNAP-19 program; and Savannah River Laboratory, operated by E. I. du Pont de Nemours & Company for the AEC, prepared the raw plutonium fuel.

Two 15-watt ^{238}Pu-fueled heaters are expected to be used to prevent instrument freezing in the first manned lunar landing mission. With these heaters the work load of the rocket will be less than with the 50-watt SNAP-27 generator considered earlier.

Contract with Nuclear Materials and Equipment Corporation Extended

The AEC has exercised an option to extend for an additional year its contract with the Nuclear Materials and Equipment Corporation (NUMEC) for operation of the Commission's boron-isotope-separation plant near Niagara Falls, N. Y., according to S. R. Sapirie, Manager of the AEC's Oak Ridge Operations. Annual expenditures under the contract are approximately $2 million. NUMEC has operated the boron plant since 1964, at which time the facility was reactivated, after being in standby since 1958, because ^{10}B inventories were not sufficient to meet future demands for all AEC programs.

Radioisotopic Heart-Assist Contract

Hittman Associates, Inc., has been selected by the AEC to design, develop, test, and evaluate an iostopic heat source for use in heart-assist devices. The 1-year contract is for $250 thousand.

XUM

Transportable ^{252}Cf Neutron-Radiography Source

A program has been initiated at Battelle–Northwest, under sponsorship of the Division of Isotopes Development, to develop a transportable neutron source for neutron radiography, using ^{252}Cf. To date a 260-μg ^{252}Cf source has been obtained and a prototype apparatus built. A complete computer-assisted analysis of various moderator and imaging materials has been performed to determine optimum conditions for thermal-neutron radiography, and a preliminary computer evaluation of materials suitable for imaging fast neutrons for fast-neutron radiography has been completed. Experimental work is under way to evaluate the prototype system as a tool for neutron radiography. (MG)

1970 Isotopes in Hydrology Conference

The AEC will coordinate the U. S. participation in a Symposium on the Use of Isotopes in Hydrology to be convened by the International Atomic Energy Agency in Vienna, March 9–13, 1970. Persons wishing to contribute papers should submit abstracts, no later than Aug. 4, 1969, to John H. Kane, Special Assistant for Conferences, Division of Technical Information, USAEC, Washington, D. C. 20545.

General

Turkish Grain Irradiator Abandoned

The Turkish government, for political reasons, has refused to license the IAEA- and UKAEA-sponsored grain-irradiation plant in Iskenderun. Several possible dispositions of both the cobalt and the plant itself have been suggested, but definite action has not yet been taken.

Limited Food Irradiation Considered in Britain

Britain's Food and Health Ministry has proposed some changes in the regulations concerning irradiated food to permit irradiation sterilization of diets for organ-transplant recipients. The proposal was to become law on March 31 unless valid objections were raised.

Radioisotope-Powered Heaters for Moon Landing

Monsanto (Mound Laboratory) has designed and fabricated two heaters fueled with ^{238}Pu ceramic microspheres to be used on the first moon landing to keep the Passive Seismic Experimental Package within its operating temperature range. The heaters will provide 15 watts(t) for a 2-year experiment life.

Large WPC Contract Awarded

American Novawood has announced that it has received a $1.2 million order from Del E. Webb Construction Company to supply 10 acres of irradiated wood–plastic composite (WPC) parquet flooring as well as guard rails for the new Kansas City International Airport terminal. The American Novawood product is sold under the tradename Gammapar.

Idaho Nuclear Energy Commission

The Idaho Nuclear Energy Commission (INEC) was established July 1, 1967, by the Idaho State Legislature, through a bill that provides for the appointment of five Commissioners as well as the establishment of an Office of Nuclear Energy Development and the appointment of an Executive Director for that office.

The INEC catalyzes, coordinates, and communicates with other structured and funded organizations in the fields of research, education, agriculture, mining, industrial expansion, and environmental development in order to advance the nuclear possibilities in the state. The Office of Nuclear Energy Development, headed by Gene P. Rutledge,* was formally opened in Idaho Falls on Sept. 20, 1967.

There is a close cooperation between the INEC and various State, Regional, and National Offices: the State Departments of Health and of Commerce and

*Formerly with Union Carbide Nuclear (Oak Ridge, K-25), Goodyear Atomic (Portsmouth), Westinghouse Electric (Pittsburgh and Idaho Falls, Bettis); prior to accepting his present position, he managed the test programs associated with the *Nautilus* prototype reactor facility S1W at the National Reactor Testing Station.

Governor Don Samuelson (left) and Executive Director Gene P. Rutledge (right) review legislation associated with the Western Interstate Nuclear Compact. Governor Samuelson has been a strong supporter of Idaho's nuclear programs.

Development, the State Water Resources and Land Boards, as well as the Attorney General's and Governor's offices; the Federation of Rocky Mountain States and the Committee on Western Interstate Nuclear Compact; Idaho's Congressional Delegation; and the U. S. Atomic Energy Commission. The INEC helped foster establishment of the Eastern Idaho Nuclear Industrial Council—a community-type organization interested in promoting applications of nuclear energy. A special interplay exists between the INEC and the Nuclear Science and Engineering Department at Idaho State University. Professor A. E. Wilson is not only responsible for the university nuclear program but is also a member of the INEC.

The INEC also urged the signing of an atomic energy agreement with AEC, which gives Idaho regulation of private uses of radioactive sources. Idaho is the 19th such agreement state, with the agreement having become effective Oct. 1, 1968. More recently, in order to provide a regional nuclear contact, the INEC has been active in its support of a plan to establish a Western Interstate Nuclear Pact, somewhat similar in operation to the highly successful Southern Interstate Nuclear Board.

The INEC is actively supporting an effort which has the goal of establishing a Nuclear Science and Information Center for the West in Idaho Falls. An excellent location on about 7 acres of land has been identified.

Of particular current significance is the interest of the INEC in getting the Materials Testing Reactor (MTR) redesignated the Western Beam Research Reactor. This effort recently culminated in the state earmarking $200,000 for nuclear oriented research associated with this facility. The INEC has worked closely with the Rocky Mountain Science Council, the Federation of Rocky Mountain States, the Western Governors' Conference, and the Western Council on State Governments in drafting a resolution requesting that the U. S. Atomic Energy Commission retain the MTR as a regional research facility, since it is the outstanding reactor west of the Mississippi. It will provide a center for beam-neutron research, accessible to western universities, and will contribute significantly to development of education and nuclear research programs in the region. It also will help considerably in establishing the State of Idaho as a nuclear research and industrial center.

XUM

The INEC, through the Office of Nuclear Energy Development, is quite active in an educational program designed to advance the nuclear possibilities of the state by stimulating interest in and knowledge of the state's nuclear resources and opportunities. These efforts, carried out with the help of the AEC's Idaho Operations Office, the Oak Ridge Associated Universities, the American Nuclear Society, and the State Chamber of Commerce as well as various local Chambers, have included:

1. Bringing the AEC's lecture–demonstration, "This Atomic World," to the Western Idaho Fair for a week; and presenting a "Nuclear Energy in Space" display at the Eastern Idaho State Fair.
2. Setting up a modular nuclear display in the Idaho Statehouse in Boise.
3. Promoting nuclear industrial seminars to cover various phases of nuclear applications and the nuclear industry.
   a. Nuclear fuels.
   b. Process radiation.
   c. Nuclear power future.
   d. Waste management.
   e. Fuel reprocessing.
   f. Plowshare.
   g. Food irradiation.
   h. Tracing and gaging with isotopes.
4. A significant demonstration of fish irradiation at the famous Snake River Trout Farm in Buhl, Idaho. (Important potato-irradiation tests are now under way at the University of Idaho experiment station in Aberdeen.)
5. A Thorium Information Meeting in Salmon, Idaho, in late 1968; a booklet on these transactions was released Apr. 14, 1969.

Idaho has considerable mineral resources, including uranium, and possesses the third largest supply of thorium in the world. Hence, the state is very much interested in development and utilization of these resources and, on Mar. 12, 1968, approved a policy of granting certain mineral leases provided the holder would "actively and vigorously prosecute and follow through on exploration." As a result huge tracts of state land have been leased to a number of major companies for exploration, with branch offices established in Boise. (PSB)

IAEA Sponsors *Engineering Compendium on Radiation Shielding*

Engineering Compendium on Radiation Shielding: Volume I, Shielding Fundamentals and Methods, R. G. Jaeger, E. P. Blizard, A. B. Chilton, M. Grotenhuis, A. Hönig, Th. A. Jaeger, and H. H. Eisenlohr (Eds.), Springer-Verlag New York, Inc., 1968, 537 + xii pages, 467 figures. $60.

The first volume of *Engineering Compendium on Radiation Shielding*, entitled *Shielding Fundamentals and Methods*, has recently become available. To satisfy the need of engineers faced with problems of shielding design, this compendium has been published under the sponsorship of the International Atomic Energy Agency as an authoritative and up-to-date summary of nuclear radiation shielding technology which includes contributions by more than one hundred outstanding specialists in this field. It covers the theoretical and experimental results of shielding research workers in almost every country having nuclear energy programs.

In addition to reviewing the fundamentals underlying the shielding of gamma rays and neutrons, it provides an understanding of the approach to the shielding problems presented by nuclear reactors, hot cells, particle accelerators, radioisotopes, and fission-product deposition from nuclear explosions. There is also a full discussion of many of the practical problems inherent in shield design: shield heating, effects of ducts and voids, and the choice of materials.

This first volume comprises eight Chapters which cover (1) Dosimetric Fundamentals and Irradiation Limits; (2) Radiation Sources (radioactive sources, particle accelerators, and reactors); (3) Radiation Attenuation Methods (Monte Carlo, point kernel, and transport); (4) Photon Attenuation; (5) Neutron Attenuation; (6) Extended Radiation Sources (point kernel integrations); (7) Radiation Induced Heat Generation (including, among other items, a survey of methods for calculating gamma-ray heating and consideration of heat generation by neutrons); and (8) Ducts and Voids in Shields.

Wherever appropriate, the authors review basic concepts and calculational techniques, compile the necessary data for shielding calculations, and offer examples to illustrate the methods described. All in all, it represents what is probably the most complete presentation of the subject currently available in a single publication. (PSB)

Radiation Technology, Inc., Plans New Irradiation Facilities

Radiation Technology, Inc., of Long Island City, New York, has leased the St. Hilaire, Quebec, irradiation facility designed for potato irradiation. The company has installed a new automatic ^{60}Co irradia-

tor system within the existing cell, which allows them to efficiently process packaged materials such as chemicals and medical disposables. The original hardware is used for low-dose bulk processing. An additional irradiation facility is planned for the Greater New York City area and will house electron accelerators and an automatic ^{60}Co irradiator. The company's principal areas of activity at present are emulsion polymerization, organo-metallic synthesis, and plastic impregnation of wood and concrete. (MG)

Moleculon Expands in Radiation Developments

Moleculon Research Corp., 139 Main Street, Cambridge, Mass. 02142, has recently received 10 new contract awards, totaling more than $300 thousand, from government and industry. The contracts all relate to radiation effects on plastics and other materials. Dr. Arthur Obermayer, President, stated that new commercial applications of radiation include upgrading of wire insulation by radiation cross-linking; finishing of textiles to impart flame-retardancy, permanent-press, or stain-release properties; and curing of paints and coatings.

During the initial 8 years of operation, Moleculon's activities were primarily in government contract research in the radiation field. But the company is now rapidly expanding its radiation processing activities in product areas, with particular emphasis on flame retardants and use of radiation in improving the properties of many common plastics.

Survey on Standard Materials

The National Research Council, through its Subcommittee on the Use of Radioactivity Standards, has initiated a survey of status of and needs for standard radioactive materials. A questionnaire has been prepared to aid in obtaining the information needed in the survey. Anyone having present or anticipated needs for standards of nuclides not now available, or of a form, strength, or accuracy not obtainable, may obtain a questionnaire from

Mr. S. A. Reynolds
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, Tenn. 37830

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Announcing the publication of

LATE SOMATIC EFFECTS OF IONIZING RADIATION by C. D. Van Cleave

This book is a compilation of what is known about the late effects of radiation at the various levels of biological organization. It is directed primarily to the nonspecialist.

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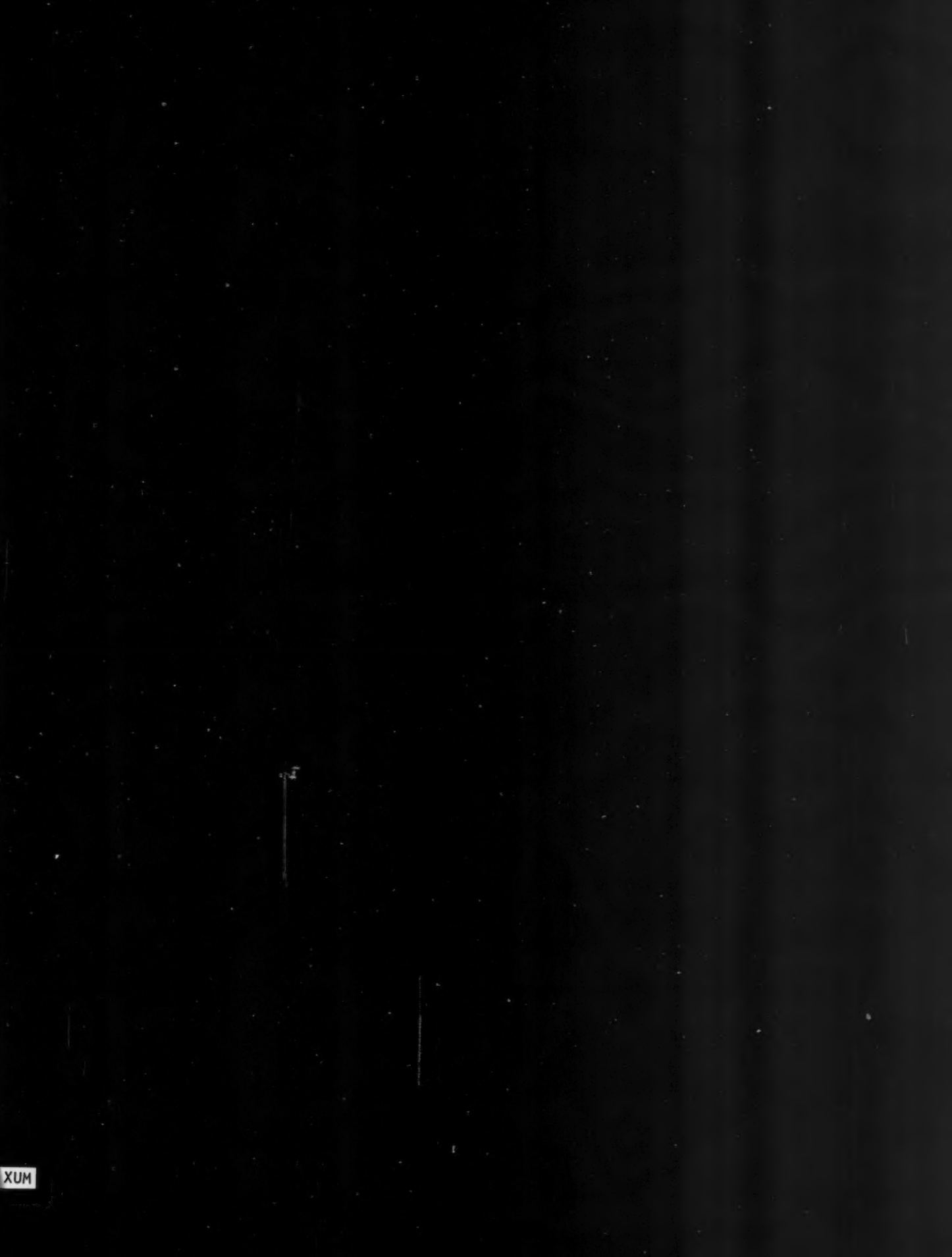
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